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A STUDY OF ABSORPTION BY LIQUID DROPS

A THESIS

Presented to

the Faculty of the Graduate Division

Georgia Institute of Technology

In Partial Fulfillment

of the Requirements for the Degree

Doctor of Philosophy

in the School of Chemical Engineering

By

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September 1956

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A STUDY OF ABSORPTION BY LIQUID DROPS

Approved:

[redacted]

Date Approved by Chairman: Sept. 7, 1956

ACKNOWLEDGMENTS

The author is grateful for the inspiration and encouragement offered by his advisor, Dr. William Meese Newton, during the course of his graduate studies.

He graciously thanks the Humble Oil Company for providing the fellowship which he was granted.

He gratefully recognizes the work of the many "Special Problem" students in the School of Chemical Engineering who have done much to lessen the task of the author.

Last, but not least, he wishes to acknowledge the faith and patience shown by his wife, Betty, and his sons, Tony and Drew.

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CHAPTER I

INTRODUCTION

General.--Gas absorption is one of the basic chemical engineering unit operations. It involves the process by which one or more soluble components of a gaseous mixture are dissolved in a liquid.

The apparatus used for contacting a liquid and a gas stream continuously may be either a tower filled with packing material, a wetted wall column, a spray column, or a plate type column containing bubble cap or sieve plates. Usually the gas and liquid streams are caused to flow countercurrently past each other through the equipment in order to attain the maximum rate of absorption.

Gas absorption processes are widely used in industry today. This process is employed over a range of applications varying from the recovery of valuable process products to the elimination of obnoxious process by-products. The petroleum industry in particular depends to a large extent upon various forms of the process in the refining and production of the hundreds of petroleum products and petrochemicals. In comparison, the roasting process, whereby copper and zinc ores sometimes containing sulfides are converted to the metallic oxides, presents an example where an obnoxious process by-product must be removed to prevent an air pollution and consequent health hazard from being created in the vicinity. Other examples are the manufacture of hydrochloric and sulfuric acids where hydrogen chloride or sulfur trioxide gases are absorbed by water or acid solutions to form the solutions commonly marketed.

Frequently the substance or substances to be recovered or removed from an industrial process appears as one or more of the components of a gaseous mixture passing through the system. The rate of absorption is determined by consideration of possible absorbents, flow rates to be encountered, equipment available, type of separation process to be used, and so forth. Adsorption, absorption, distillation and extraction are probably the most widely utilized and accepted processes used to remove or separate constituents from a mixture.

All of these processes involve essentially the same mechanism of mass transfer from one phase to another. If a complete study were made of the primary mechanisms of mass transfer for one type of process, for example, absorption, the basic ideas might well be applied to similar studies conducted upon the other mass transfer processes.

While chemical engineering unit operations have been practiced in one form or another since the earliest days of chemistry, there still remains much to be learned about the theory. It has only been in the past century and more specifically in the past fifty years that a very definite attempt has been made to assimilate and correlate the numerous studies on unit operations. As a result of this compounding of knowledge, most of the questions involving basic fundamentals of chemical engineering unit operations have been answered to some extent. Current investigations have become more complex due to the increasing number of variables considered. Today a larger amount of attention is devoted to the problem of reducing the results of complex studies to some formulation based upon the fundamental principles governing the system investigated.

The field of gas absorption, however, does not appear to have

reached this stage of development. Investigators of gas absorption phenomena have presented a range of results which vary as widely as the range of variables encountered. The basic principles of gas absorption are founded upon a relatively small number of investigations made twenty-five to thirty-five years ago. Since these early investigations, few studies have been made in the gas absorption field either to reaffirm the results of the original studies or to further investigate the fundamentals of gas absorption. Attention has been directed to the study of the effects of various variables upon the rate of gas absorption, ignoring completely the establishment of a firm foundation of basic principles.

Gas absorption.--Absorption may be accomplished in either a continuous or a batchwise process. Some investigators (1,2,3,4) who have studied batchwise processes have concluded that the rate of absorption under these conditions is a function of pressure, temperature, the nature of the solute and solvent, and the area of contact. Also Becker (2) and Hutchings (4) report that the rate of absorption is a function of agitation, that is the rate of absorption may be increased by mechanical stirring which, it is postulated, increases the turbulence and thus serves to the decrease the over-all resistance to absorption.

O'Brien and Stultzman (1) investigated the case of the evaporation of various liquids by a turbulent air stream passing over a free and more or less smooth liquid surface. The results of this study were evaluated and an attempt was made to correlate them with the results obtained by other investigators (5,6,7,8) using the same liquids under approximately the same conditions of pressure and temperature in both wetted wall and packed columns.

The batchwise process, while interesting in all phases of unit operations, has been displaced in most investigational studies by the continuous process for some of the same reasons that it has been replaced in industrial processes - ease of control and operation.

The emphasis in industry upon increased equipment capacity has necessitated the trend of absorption systems from the initial spray column to the plate column and now at present the packed column due to the increased amount of surface area available for mass transfer presented by each. The majority of the investigations of gas absorption at present are centered upon the packed column system. Investigators have studied such problems as the effect of liquid hold-up in the column, types of packing materials involving shapes and surface area per unit weight, height of packing, pressure drops in packed columns, flooding rates, packing diameter and column diameter, and many others. These investigations brought with them the need for new terminology in gas absorption and in fact an entirely new method for expressing the rate of gas absorption. With the use of packing materials, it became impossible to effectively evaluate an expression for the area available for mass transfer in a packed column. Thus the absorption coefficient and the area are usually reported as a single group. The problem of the evaluation of experimental results was made more difficult by this step since two variables were combined into one experimentally determined value. The studies of packed columns led also to the introduction of such terms as the number of transfer units (N.T.U.) and the height equivalent to one theoretical plate (H.E.T.P.).

Wetted wall columns offer more opportunity for absorption studies

in so far as the available area for mass transfer is easier to evaluate. In a wetted wall column the liquid is introduced to the column wall at the top of the column and is allowed to flow down the wall in streamline flow, that is the Reynold's number of the liquid flow is in the laminar region. Some investigators (9) report however that turbulent flow of the wall liquid occurs in a wetted wall column at Reynold's numbers as low as 1,600 to 2,000. This determination was made with the use of a stationary air core in the column, i.e., no gas flow, which led the authors to postulate that turbulence could be obtained in the wall liquid at even lower Reynold's numbers if the gas rates were sufficiently high to disturb a laminar wall liquid flow pattern, if any.

The literature dealing with spray columns constitutes a small fraction of the total number of absorption studies. This may be attributed to the larger number of operational variables encountered. The major problem in spray columns is how to achieve efficient contact between a gas and the liquid spray. If this is to be a countercurrent operation, the problem becomes more difficult because to achieve an efficient spray the liquid must be sprayed under pressure through a commercial type spray nozzle or similar distributing device. If the drops are too small they are slowed down by air friction and possibly carried out of the column by the exit gas as entrained liquid. If the drops are too large they tend to strike the walls of the column due to the circular effect of most nozzles and consequently effect a decrease in the area available for mass transfer.

Johnston and Williams (10) estimated the interfacial area present in a spray chamber into which liquid drops of the same size as those

produced by a commercial spray nozzle were injected vertically downward along lines parallel to the axis of countercurrent air flow. In this idealized situation in which it was assumed that no liquid struct or adhered to the wall, it was found that the interfacial area of the spray per unit tower volume decreased with increasing distance from the nozzle.

Pigford and Pyle (11) investigated quite extensively the humidification of air, stripping of oxygen from water, and the absorption of ammonia by water in 26 and 52 inch high, 31.5 inch diameter spray columns utilizing six number 5-B Sprayco solid cone commercial spray nozzles. They observed that despite the fact that the nozzles were arranged so as to produce a spray directed toward the center of the column, the water running down the walls of the column amounted to between 60 and 80 per cent of the total liquid sprayed. They provided the system with a take-off device which separated the spray core liquid from the wall liquid and proceeded to analyze both liquid streams. They concluded that the rate of interfacial transfer at the drop surface is most rapid just after the drops are formed at the nozzle. It was also noted that the interfacial area of the spray was about in propostion to the volume of spray present in the chamber at any instant, the increased effect of coalescence of the drops at higher spray pressures being offset by the smaller initial drop size. It was observed that spray chambers are reasonably efficient for an operation in which the liquid phase predominates by comparison to a duplicate chamber filled with packing material and operated under the same flow conditions. In the same comparison it was also noted that an increase in the height of the packed column would increase the efficiency of that unit more than a similar increase in the

height of the spray chamber since appreciable countercurrent action evidently does not take place in a spray column since the gas appeared so thoroughly mixed so as to assume it is nearly the same composition throughout the entire column volume.

Hixon and Scott (12) studied the relations between tower heights and various gas and liquid flow rates for the absorption of ammonia and sulfur dioxide by water and the absorption of benzene vapor by straw oil in a spray type column. They concluded that the over-all coefficient of absorption based on a wall free inner cone section of the tower is proportional not only to the gas flow rate to the 0.8 power but also the liquid flow rate and inversely to the tower height.

Additional investigations (13,14,15,16) on spray column operation are of a limited nature dealing with such variables as types of spray nozzles, liquid and gas flow rates, and column dimensions. They are mentioned briefly to summarize the work done in the spray column field.

It is thus apparent that in order to make a comprehensive study of the basic principles of gas absorption the investigation must be in a field where it is possible to limit the number of variables to a minimum. For gas absorption this appears to be the general field of the absorption by liquid drops, since in this case the interfacial area is subject to control. Unfortunately only a small number of studies have been conducted in this field and the majority are of such a limited nature and differ so widely in investigational techniques that it is not possible to present an adequate comparison or correlation of the reported results.

Whitman, Long, and Wang (17) presented the first investigation

on the absorption of gases by single drops falling through a column of fixed height with varying rates of drop formation. They studied the rate of absorption obtained for various column heights and then extrapolated to the zero time of formation to determine the amount of absorption which took place during the time of fall.

Hatta, Ueda, and Baba (18) made a similar study with towers of varying heights. No data was presented as to the absorption taking place during the time of drop formation. Both investigations (17,18) however do indicate the existence of high coefficients of absorption for freely falling drops.

Froessling (19) presented some measurements on the rate of evaporation of drops of water, aniline, and nitrobenzene by an air current. The drops were suspended on a thin glass rod and the rate of evaporation was measured photographically. The resulting data was correlated by an empirical equation on the basis of dimensionless groups containing the velocity of air, the size of the drop, and the properties of the diffusing liquids.

Johnstone and Williams (20) investigated the absorption of sulfur dioxide, carbon dioxide, hydrogen chloride, hydrogen sulfide, and ammonia by various acids or bases. They also studied the deformation of drops and the time of fall of various sized drops experimentally and theoretically. They concluded that drop deformation effects and variations in fall time due to various gas velocities were negligible.

Shabalin (21) studied the absorption of ammonia and carbon dioxide by liquid drops of various sizes in an apparatus so constructed as to give a constant velocity to the drop through the absorption zone. He

concluded that the absorption of a gas by liquid drops of small diameters was analogous to heat transfer and depends thus on the diffusion of the gas into the immobile interior of the drop.

Hatta and Babba (22,23) investigated the absorption of carbon dioxide by water drops. They developed three theoretical relations for expressing the condition of the surface of a free falling drop. The first condition postulated the presence of turbulence in the surface of the drop during the time of formation, the second a development of turbulence in the drop surface during the fall of the drop, and third, no turbulence at all. The results of their studies led them to conclude that the last postulate was correct. They also determined the amount of carbon dioxide absorbed during the formation of the drop and concluded that this absorption was a negligible amount. Finally they observed that most of the absorption took place during the initial stage of the fall and concluded that it would be better to increase the frequency of the drops than to extend the fall distance.

It has been shown that mass transfer in a gas-liquid system depends on diffusion through films. Most investigators have studied mass transfer while the gas phase was in turbulent motion and assumed that the liquid film had no resistance. In the case where the liquid film must be considered, the chemical nature of the liquid, liquid flow rate, liquid viscosity, density, and surface tension may be factors which affect absorption or desorption and should be considered.

Effect of surface active agents upon absorption.--Riou (24) in a study of the rates of absorption of carbon dioxide gas in a water solution of

sodium carbonate in a packed column noted that the addition of surface active agents such as ethyl glycol, methanol, lactose, formaldehyde, peptone, pepsine, and soluble gelatin produced decided effects. The effects, however, varied over the range of producing an increase in the absorption rate of carbon dioxide to an observed decrease in the absorption rate. Riou stated that the viscosity of the solutions should remain essentially unaltered by the relatively small amounts of the agents added. In essence he postulated finally that the effects which he had observed were due to catalytic surface reactions.

Killefer (25) reinvestigated the overall studies of Riou (24) utilizing formaldehyde, phenol, sodium lactate, and ethanolamine as surface agents. He found that the rate of absorption was increased greatly in packed column. This effect led to the hypothesis that the change in the absorption rate was due to the lowering of the surface tension of the solutions and was not related in any manner to a catalytic action of the added surface active agents.

Rennolds (26) observed after a series of studies on the desorption of carbon dioxide from water in a packed column that the rate of desorption was lower when a surface active agent was employed than the rate of desorption obtained with plain water under the same flow conditions. The agents used were W-70 (manufactured by Dewey and Almy), Ultrawet (Atlantic Refining Company) and formaldehyde. He concluded that this effect was observed only in the case where the liquid film was the controlling film. These results are substantiated by other investigators (27,28,29,30,31,32,33) studying the evaporation of various liquids by air in the presence of mono-or-multi-molecular films.

Sherwood (34,35,36) performed similar experiments to those of

Rennold's and stated that the addition of surface active agents affects the absorption rate when the liquid layer or film is controlling and that the added agents have no effect upon the absorption rate when the gas film is the principle resistance to diffusion. Sherwood further postulates that the decrease in the rate of absorption is due to the presence of the large molecules of the surface active agent upon the liquid surface which tend to impede the movement of the diffusing molecules through the liquid film.

Pozin (37,38) studied the absorption of ammonia and carbon dioxide by various acid and alkaline solutions. He concluded that the rate of absorption was in no way related to the surface tension of the absorbing liquid solutions.

Trenovoskaya and Belopolskii (39) have presented the most recent and thorough investigation of this topic. Their studies were based upon the absorption of sulfur dioxide by water solutions in a wetted wall column utilizing such surface active agents as "merzolyat" (a mixture of sodium salts of sulfonic acids and hydrocarbons boiling between 230° and 240° Centigrade), "nekale" (sodium salts of isobutyl-B-naphthalene sulfonic acid), and "sulfonol" (a mixture of sodium salts of sulfonic acids and hydrocarbons from kerosene). They observed that small amounts of these agents in the absorbing water phase produced a definite decrease in the absorption rate. They observed also that as the concentration of the added agents were increased the absorption rate continued to decrease until a minimum was reached which was in the range of 25 to 35 per cent less than the absorption rate obtained with the use of plain water under the same operating conditions. Their investigation revealed that the

decrease in the absorption rate ceased at very low concentrations before the so-called inactive portion of the concentration versus surface tension isotherm was obtained, that is, that portion of the surface tension curve where a further increase of the added agent concentration produces little or no change in the surface tension of the solution. This led them to conclude that the change in the absorption rate could not be attributed to the change in the surface tension of the solution and that the answer must therefore lie in the formation of a layer of the surface active agent molecules upon the surface of the liquid solution. They also observed a change in the flow pattern of the water flowing down the wetted wall column upon the addition of small amounts of the surface active agents. In a further discussion of this investigation (40) the Gibb's adsorption equation was utilized to show that the limiting or minimum absorption rate was found at a surface active agent concentration where the Gibb's equation predicted a maximum excess surface concentration. The results obtained through the use of "nekale" could not be explained by this postulation and the idea was advanced that this might be due to the fact that the structure of the liquid surface film was altered through the reorientation of the surface active agent molecules.

Johnstone (41) found that the addition of small amounts of Tergitol decreased the extraction of acetic acid from isopropyl ether by water drops in a packed liquid extraction column to one third of that obtained in the absence of Tergitol. This he postulated was due to a change in the interfacial tension of the liquids.

Chu, Taylor, and Levy (42) observed that the rate of extraction in a packed column increases linearly with decrease in interfacial

tension caused by the addition of small amounts of surface agents. They concluded, however, that this was because the packing was able to break up the disperse phase into smaller drops and consequently increase the surface area.

Garner and Skelland (43) investigated the effects of specific agents of known structure and of anionic, cationic, and non-ionic types for a system which involved the transfer of acetic acid from nitrobenzene droplets to water. They concluded that the rate of mass transfer depended upon both, the structure of the molecule and the type of agent and postulated that several different types of static film structures may be produced which could alter the observed effects upon mass transfer in the system under study.

Structure of surface films.--It is in order at this point to briefly outline the presently accepted theories on surface films and their structure in order to develop some definite thoughts to be used later in the analysis and evaluation of the experimental data obtained through the course of this investigation.

The basic relation used to determine the concentration of a surface active agent in the liquid surface is the adsorption equation first derived by J. W. Gibbs (44) and presented in detailed discussion in Chapter II - Theory.

Langmuir (45) concluded from his studies of films of insoluble substances upon water that the film formed was but one molecule thick and that the molecules in the liquid film were definitely oriented. He advanced the theory that in such cases for both soluble and insoluble

substances and for pure liquids the Gibbs film may be regarded as but one molecule thick and consisting of pure solute molecules which lower the surface tension of the solution. This is in agreement with the first published work on the subject by Lord Rayleigh (46) who initially postulated surface films on water as consisting of a single layer of molecules which could be regarded as floating objects. The work of Langmuir has been substantiated by Harkins (47,48) and others and today is recognized as one of the basic principles of surface chemistry.

Rideal (49) states that in mixtures of substances of markedly different surface tensions it has been found that over a considerable range of concentration the Gibbs film appears to behave as if it were unimolecular in character, but for strong solutions of these substances as well as for mixtures of liquids of similar surface activities the evidence is by no means conclusive. It is further stated that it must be assumed that in these cases the application of the principle of minimum surface energy to mixtures somewhat similar in internal attractive forces leads to the formation of a diffuse layer in which the concentration varies possibly in an exponential manner with the depth, the top layer alone may be said to be formed by the operation of chemical forces. This led Rideal to postulate for the case of concentrated solution of ethyl alcohol in water, that the subsequent decrease in the excess surface concentration with the increase of the concentration of the alcohol in the liquid bulk might, on the assumption of an oriented layer of alcohol molecules in the Gibbs film, be accounted for by the decrease in the orientation of the surface molecules when the more polar water molecules in the bulk phase are displaced or replaced by the alcohol molecules.

Rideal states further that although most mass interphase transfer takes place at great speed as in the vaporization and condensation of water at the normal boiling point, the life of a molecule on the surface is probably long enough to permit such adjustment as orientation requires.

Young and Coons (50) state that due to the presence of unbalanced forces on a liquid surface and the apparent effort of all liquids to display a minimum surface, the surface of a liquid has very different characteristics from those of the interior. They discussed the case of a spherical drop of liquid whose surface is described as from one to three layers of molecules thick. The remainder of the molecules in the drop are thus situated in a region of unbalanced force, that is the attractive force exerted on a molecule in the liquid bulk by the molecules in the liquid bulk is greater than the attractive force exerted on this same molecule by the surface molecules. Thus a molecule in this region tends to remain in the liquid bulk. The horizontal forces are in balance, but due to the unbalanced force of attraction exerted by the bulk molecules the surface molecules tend to move toward the center of the drop until all of the forces are thus in balance. At the same time, there is a lateral movement of the surface molecules to fill the space left vacant by the molecules which have been pulled into the interior of the sphere. At some places, other molecules are moving by translational movement into the surface layer. If these movements are added to the usual activity at a surface when mass transfer is in progress, the average life of a molecule is very short. It is estimated here that the average life of a molecule in the surface layer is less than one-fifty millionth of a second.

Investigations by Hedestrand (51) indicate that a mono-molecular film of oleic or palmitic acid upon water does not diminish the rate at which water evaporates into a current of air moving over it.

Harkins and Morgan (52) concur with this finding in a study of the evaporation of water covered by a mono-molecular film into still air. These results led them to conclude that the mono-molecular films contain water as well as the organic substance and, further, that the film is anything but a static structure.

The manufacturers of Aersol OT (53), one of the agents utilized for this investigation, state that tests have indicated that the agent has no effect upon the rate of evaporation of water solutions containing various concentrations of the agent.

Harkins (54) stated that aqueous solutions of most soaps exhibit a more or less abrupt change in physical properties over a relatively short concentration range and that this phenomenon has been attributed to the formation of oriented soap aggregates. These changes in physical properties have been observed in studies on freezing point depression (55), density (56), viscosity (57), surface tension-time effects (58) and others.

Vapor pressure and drop size.--Small droplets must have a higher vapor pressure than an extended surface, the difference depending upon the radius of the drop. Eucken (59) and co-authors present a derivation of the relation between vapor pressure and drop radius based upon the relation between mass and surface area and the second law. It is then shown by actual calculation that deviations in the vapor pressure of a liquid drop are not encountered until a droplet radius of approximately 10^{-5}

centimeters is encountered.

Surface tension and drop size.--Tolman (60,61,62) presented several studies of the effect of droplet size on surface tension through utilization of the Gibbs surface theory and concluded that the effect of radius of curvature on surface tension would be small until very small radii were reached.

Keenan (63) also concluded that the surface tension of a liquid can be expected to decrease with a decrease in radii.

Kirkwood (64) developed a general statistical mechanical theory of interfacial phenomena and presented expressions relating surface tension and other thermodynamic functions to the potential of intermolecular force and molecular distribution functions. The derived expressions were then applied to liquid Argon at 90° Kelvin and compared with experimental values. This led to the conclusion that the quasi-thermodynamical theory expressed by Tolman (60,61,62) was correct and that the surface tension of small drops decreases with increasing curvature.

These results have also been reported by Koenig (65) and Hill (66) using entirely different methods of approach to the problem.

The effect of surface tension on droplet size does not make itself apparent until the drops are of the order of 20°A in diameter.

Purpose of this investigation.--Various investigators have employed a variety of surface agents in absorption studies. The amounts of the agent used were usually small, which led to the supposition that the two important liquid properties, density and viscosity, were unchanged. These studies have also produced a variety of reported results. No

attempt has been made to relate surface tension and surface agent concentration to these results. Some investigators claim that the lowering of the surface tension of the absorbing liquid by the addition of a small amount of a surface active agent decreases the rate of absorption, while another group claims that the absorption rate is increased by lowering the surface tension. Still a third group has asserted that surface tension has no relation to the observed phenomena. All investigations have been hindered by the unknown and usually unconsidered variable of surface available for mass transfer.

The purpose of this investigation was to make a study of the various factors which affect the absorption of gases by liquid drops, with particular emphasis upon the surface available for mass transfer and the effect of the addition of surface active agents to the liquid phase. Water was chosen as the absorbent for three gases, carbon dioxide, sulfur dioxide, and ammonia, which offer a wide range of solubility.

The scope of the investigation includes one water rate; three gases at four chosen rates and three gas compositions at each rate; and three surface active agents at seven chosen concentrations.

This study covers an investigation of the effect of gas rate, surface tension, and concentration of the diffusing gas on the absorption of carbon dioxide, sulfur dioxide, and ammonia from various air-gas mixtures by tap water in drop form, and the same systems to which various amounts of the surface active agents Aersol OT, Sterox SK, and Victawet 12 were added to the liquid phase.

CHAPTER II

THEORY

General absorption theory.--Whitman (67) first proposed the two film theory of gas absorption. He advanced the idea that two films exist at the phase boundary between two liquids or between a gas and a liquid phase. Reynolds (68) soon followed with his classical studies on fluid flow in a conduit and the theory of a stationary film at the phase boundary. Lewis and Whitman (69) proposed the hypothesis that material is transferred through these films by diffusion only and by convection in the main body of the stream. These two films may be considered as two diffusional resistances in series with an equilibrium condition existing at the interface. The difference in concentration across the films represents the potential or driving force that causes the mass transfer. From diffusional principles and the law of continuity for steady state transfer, which states that any material passing through one film must also pass through the interface and the other film, the rate of diffusion may then be expressed for a steady state as follows,

$$N_A = \frac{dW}{Ad} = k_G (p-p_i) = k_L (c_i-c) \qquad (1)$$

where: N_A is the rate of diffusion of the diffusing substance expressed in mass per unit area per unit time.

W is the mass or weight of the substance being transferred.

A is the effective area between the two phases.

is the time.

k_G and k_L are the film coefficients of the gas and liquid films respectively.

p and p_i are the partial pressures of the diffusing substances in the main gas stream and at the interface respectively.

c and c_i are the concentrations of the diffusing substances in the liquid stream and at the interface respectively.

In order to apply this equation, the value of k_G and k_L and the equilibrium relation between the diffusing substance and the absorbing liquid must be known. Since data on individual film coefficients are usually not available, convenient expressions employing overall coefficients, K_G and K_L were introduced so that the rate of diffusion could be calculated without a knowledge of the actual conditions at the interface. Equation (1) in terms of K_G and K_L may be written as,

$$N_A = K_G(p-p_e) = K_L(c_e-c) \qquad (2)$$

where: p_e is the partial pressure of the diffusing substance in equilibrium with a solution having a concentration c of the diffusing substance.

c_e is the concentration of the diffusing substance in a solution in equilibrium with the diffusing substance in the vapor state with a partial pressure of p.

If the equilibrium relation follows Henry's law, then the overall coefficient and the individual film coefficients are related by the equation,

$$\frac{1}{K_L} = \frac{1}{HK_G} = \frac{1}{k_L} \neq \frac{1}{Hk_G} \qquad (3)$$

where H is Henry's law constant.

Mass transfer to or from a sphere placed in a stagnant fluid, for example, the evaporation of a liquid drop in absolutely still air, follows the laws of molecular diffusion. Langmuir (70) has shown that the radial diffusion into a quiet medium of infinite size may be expressed by,

$$N_A = \frac{2D_v(p_i - p)}{DRT} \qquad (4)$$

or

$$\frac{k_c D}{D_v} = \frac{k_G RTD}{D_v} = 2 \qquad (5)$$

where: D is the diameter of the sphere.

p_i is the vapor pressure at the surface.

p is the partial pressure of the diffusing material at a remote point (theoretically at infinity).

D_v is the molecular diffusivity in the gas.

R is the gas constant.

T is the temperature.

N_A is the rate of mass transfer.

k_G is the individual or gas film coefficient.

k_c is $k_G RT$, the mass transfer coefficient.

Numerous investigators (19,71,72,73,74) have definitely established

this relation and have found that the value of $k_c D/Dv$ approaches 2 as an asymptote as the velocity of the fluid medium approaches zero.

Froessling (19) reported extensive data on the evaporation of small drops of nitrobenzene, aniline, and water and small solid spheres of napthalene. He found that his data could be well correlated by the semi-theoretical equation,

$$\frac{k_c D}{D_v} = 2\left[1 + 0.276 Re^{0.5} \left(\frac{\mu}{\rho D_v}\right)^{0.33}\right] \qquad (6)$$

where: k_c is $k_G RT$, the mass transfer coefficient.

D is the diameter of the sphere.

D_v is the molecular diffusivity in the gas.

Re is the Reynolds number $Du\rho/\mu$ where:

μ is the viscosity of the gas medium.

ρ is the density of the gas medium.

D is the diameter of the sphere

u is the velocity of the drop in relation to the gas stream.

Pigford (75) noted that the effect of $\mu/\rho D_v$ was a function of the Reynold's number and developed an empirical correlation of $K_c D/D_v$ versus $(Re)(\mu/\rho D_v)^{2/3}$.

Gibbs surface theory.--A pure liquid consists of one species of molecules and the surface free energy is a linear function of the total surface. When the surface is extended, molecules must be brought from the interior of the liquid to the surface and work must be done against the interior

molecular attractive forces. To simplify the mathematical calculation of this surface energy, a hypothetical tension acting in all directions parallel to the surface equal numerically to the free surface energy is generally used. This is called surface tension.

In the case of solutions of two or more constituents whose molecules have fields of attractive force which differ in intensity, the molecules which have the greater fields of force tend to pass into the interior of the solution and those with the smaller fields remain at the surface. The surface layer of the solution will therefore be more concentrated in the constituents which have the smaller field of attractive force.

If the solution is ideal, there should be no difference between the concentration on the surface and the concentration of the interior of the liquid. Since few solutions are ideal, an excess of one constituent over the other on the surface is expected. Gibbs called the amount by which the total quantity of a component exceeds that in the idealized system as the "surface excess" and symbolized this function as Γ .

The energy of a system (E) may be expressed as a sum of terms each of which is a product of a capacity factor and an intensity factor. The capacity factors include such quantities as entrophy (S), volume (V), area (A), and the amount of the component (m); the intensity factors include such quantities as temperature (T), pressure (P), surface tension (γ), and the chemical potential (μ). For a system of two components, the change in energy of the actual system during small reversible changes at equilibrium is,

$$dE = TdS - p^{\alpha} dV^{\alpha} - p^{\beta} dV^{\beta} + \gamma\, dA + \mu_1 dm_1 + \mu_2 dm_2 \qquad (7)$$

where the superscripts α and β refer to phases, and the subscripts 1 and 2 refer to components. The change of energy of each phase of an ideal system may be written separately as,

$$dE^{\alpha} = TdS^{\alpha} - p^{\alpha} dV^{\alpha} + \mu_1 dm_1 + \mu_2 dm_2 \qquad (8)$$

$$dE^{\beta} = TdS^{\beta} - P^{\beta} dV^{\beta} + \mu_1 dm_1 + \mu_2 dm_2 \qquad (9)$$

Due to the non-ideality of an actual system the surface properties will have the following characteristics,

$$E^s = E - E^{\alpha} - E^{\beta} \qquad (10)$$

$$S^s = S - S^{\alpha} - S^{\beta} \qquad (11)$$

$$m^s = m - m^{\alpha} - m^{\beta} \qquad (12)$$

where the superscript s indicates surface.

Since there is no volume at the surface, the change in energy at the surface would be,

$$dE^s = TdS^s + \gamma\, dA + \mu_1 dm_1^s + \mu_2 dm_2^s \qquad (13)$$

which can be obtained by subtracting equations (8) and (9) from equation (7), with the aid of equations (10), (11), and (12). If the surface is allowed to increase indefinitely without a change in composition, equation (13) can then be integrated to produce,

$$E^s = TS^s + \gamma A + \mu_1 m_1{}^s + \mu_2 m_2{}^s \tag{14}$$

By differentiating equation (14), we obtain the following,

$$dE^s = TdS^s + S^s dT + \gamma\, dA + Ad\gamma + \mu_1 dm_1{}^s + m_1{}^s d\mu_1 + \mu_2 dm_2{}^s + m_2{}^s d\mu_2 \tag{15}$$

Comparing equations (13) and (15),

$$Ad\gamma = -S^s dT - m_1{}^s d\mu_1 - m_2{}^s d\mu_2 \tag{16}$$

Per unit area of the surface, equation (16) reduces to,

$$d\gamma = -S^s dT - \Gamma_1{}^s d\mu_1 - \Gamma_2{}^s d\mu_2 \tag{17}$$

At constant temperature, and with the surface so fixed that the surface excess of one of the components vanishes, equation (17) is now reduced to,

$$d\gamma = -\Gamma_1 d\mu_1 \tag{18}$$

Since by definition,

$$\mu = \mu_o + RT\ln(fN) \tag{19}$$

where μ_o is the chemical potential of some arbitrarily chosen initial standard state.

f is the fugacity.

N is the mole fraction.

Differentiating equation (19) we have,

$$d\mu = RTd\ln(fN) \tag{20}$$

Substituting equation (20) into (18),

$$d\gamma = -\Gamma_1 RT d\ln(f_1 N_1)$$

which may be written as,

$$\Gamma_1 = \frac{f_1 N_1}{RT} \cdot \frac{d\gamma}{d(f_1 N_1)} \tag{21}$$

For dilute solutions, where the concentration, c, is proportional to the mole fraction, N, and f = 1, equation (21) takes the form,

$$\Gamma_1 = \frac{-c}{RT} \cdot \frac{d\gamma}{dc} \tag{22}$$

which is the Gibbs equation for surface excess concentration.

If the surface tension of a solution of two components is measured at various concentrations, and the results are plotted as surface tension versus concentration, the slope of the curve would represent d /dc in equation (22). With the use of this value for the slope the surface excess at any concentration can then be evaluated from equation (22).

The Gibbs equation is thus based upon conditions of constant temperature and pressure in a state of static equilibrium. A further limitation is the postulation of a plane surface of negligible thickness, generally referred to as the Gibbs surface or film. The final condition is that of a dilute solution where the concentration of the solute is proportional to the mole fraction of the solute in the solution or the assumption of unit fugacity and activity.

For dilute solutions of non-ionizing or feebly ionizing solutes, no serious error is introduced by employing bulk concentrations in lieu

of activities, but in general serious errors are introduced through failure to take this precaution.

When the surface tension of the absorbing liquid in a gas-liquid absorption process is altered by the addition of a small amount of a surface active agent, the liquid surface will possess a somewhat larger amount of that agent, the concentration of which may be calculated by the method previously outlined. If this is the case, the surface active agent will then occupy a part of the interfacial area between the two phases and render the effective area through which the material transferred must pass less than in the case where no agent is utilized. In this case, the mass transfer coefficient would then depend upon the surface concentration and consequently the surface tension of the absorbing liquid.

CHAPTER III

EXPERIMENTAL EQUIPMENT

The experimental equipment consisted essentially of a glass column; a liquid reservoir; a liquid pump; an air compressor; cylinders of carbon dioxide, sulfur dioxide, and ammonia; and various devices for controlling and metering the liquid and gas streams. Schematic diagrams and photographs of the construction and layout of the experimental equipment are shown in Figures 1-8, inclusive of the main text.

The column used in this investigation was a seven and nine-sixteenth inch inside diameter, glass cylinder, four feet high, mounted in a supporting structure as shown in Figures 4 and 5. The glass column was seated with a grease seal on a plastic adaptor placed on the column bottom assembly. The grease seal was a layer of standard cup grease spread approximately one-eighth of an inch thick over one of the surfaces to be sealed. The surfaces were then joined and a grease fillet was smoothed over the sealed joint.

The air system as shown in Figures 1 and 3 was supplied with air by the central laboratory compressor at a pressure varying between 90 and 110 psig. A liquid trap was located in the line to remove any water possibly condensed in the main air supply line. The pressure was then reduced by two spring actuated diaphragm type pressure regulators in series to the approximate operating pressure of ten psig. A section of two inch standard pipe packed loosely with glass wool fiber served as an air filter to remove any small rust or dust particles or entrained water

droplets. The air stream was metered through a previously calibrated rotameter having the correct flow range within its range of metered flow. Pressure was maintained in the rotameter through control of the downstream pressure control needle valves and by adjustment of the flow rate to the rotameter by regulation of the inlet flow control needle valve. The pressure in the rotameter was indicated by a mercury manometer located on the main control panel. The by-pass loop at the downstream pressure control point was used at high gas rates where the capacity of the small control valve was exceeded. The air line then joined the additive gas line.

The additive gas stream as shown in Figures 1 and 3 was supplied by gas from a commercial gas cylinder. A pressure regulator was attached to the gas cylinder outlet and the pressure was reduced to approximately ten psig. The stainless steel finned-tube radiator allowed the gas to reach room temperature in event of any cooling during the pressure reduction stage. The additive gas stream was metered through a previously calibrated rotameter having the correct flow range within its scope of metered flow. Pressure was maintained in the rotameter through regulation of the downstream pressure control needle valve and by adjustment of the flow rate to the rotameter by regulation of the inlet control needle valve. The pressure in the manometer was indicated by a connected mercury manometer located on the main control panel. The additive gas line then joined the air line.

The air and additive gas lines were joined and the resulting mixture was passed through a system of flow restrictions to obtain proper mixing of the gases. The mixture line was provided with a control valve

to close off this portion of the system in the event of improper functioning of the column. Thermometers were located in the air, additive gas, and air-additive gas mixture lines. A separate thermometer was used to periodically check the temperature of the exit gas leaving the top of the column.

The air and additive gas mixture was introduced into the bottom of the column through the gas distributor shown in Figure 7. The distributor was constructed in a five inch square pattern from standard 3/4 inch copper tubing and fittings. Three rows of 1/32 inch diameter holes on 1/2 inch centers were drilled in the distributor, one row directly on the bottom with the other two rows being offset 1/4 inch to each side of the bottom row. The gas mixtures passed through and out of the column, which was open to the atmosphere, and were exhausted from the building by a large window fan.

A 1000 pound capacity calibrated tank served as the water reservoir. Water entered the reservoir from the laboratory water line. A recycle line was provided so that a portion or all of the liquid in the reservoir could be recycled through the pump and back to the reservoir again to mix the liquid when surface active agents were employed. A line was connected from the reservoir to a drain for convenience in draining and cleaning. The reservoir water flowed to the 1/4 horsepower pump by gravity and then was recycled or entered the water flow system. The water was then metered through a calibrated rotameter through manual operation of a needle control valve and entered the liquid distributor shown in detail in Figure 8.

The liquid distributor consisted of a four inch diameter, 1/4 inch

plexiglass plate through which were placed twenty-five 1/2 inch long, 1/4 inch brass rods in a pattern as shown in Figure 8. This pattern was designed to give the same liquid rate per unit cross sectional area over the entire distributor. The brass rods were machined round at the container end to prevent possible clogging of the holes by suspended particles settling out of the process water. One hole 0.036 inches in diameter was drilled through the center of each of the brass rods. A plexiglass cylinder four inches outside diameter and eight inches in height was then attached to the distributor plate to complete the construction of the liquid distributor.

The liquid distributor was centered at the top of the column which was in turn centered on the liquid receiver at the bottom of the column and checked for correct alignment in a perpendicular position.

The liquid receiver was a four inch outside diameter, four inch high plexiglass cup provided with a plastic tube drain line. The drain line or sample removal line was connected into the column bottom assembly as shown in Figure 6. The flow out of the liquid receiver was controlled by a stopcock which enabled a constant liquid level to be maintained in the receiver. The liquid line was then divided, providing a liquid sample take-off line and a direct drain line. Temperatures were recorded by thermometers located in the liquid entrance and exit lines.

The diagram of the column bottom assembly indicates the provisions for draining the column bottom in event of improper functioning of the column.

A barometer was installed on the control panel to provide a convenient source of barometric data.

A duNouy Interfacial Tensiometer, made by the Central Scientific Company, of Chicago, Illinois, was used to measure the surface tension of the absorbing liquid.

All exposed metal surfaces of the gas and liquid distributors were thoroughly coated with a corrosive resistant varnish.

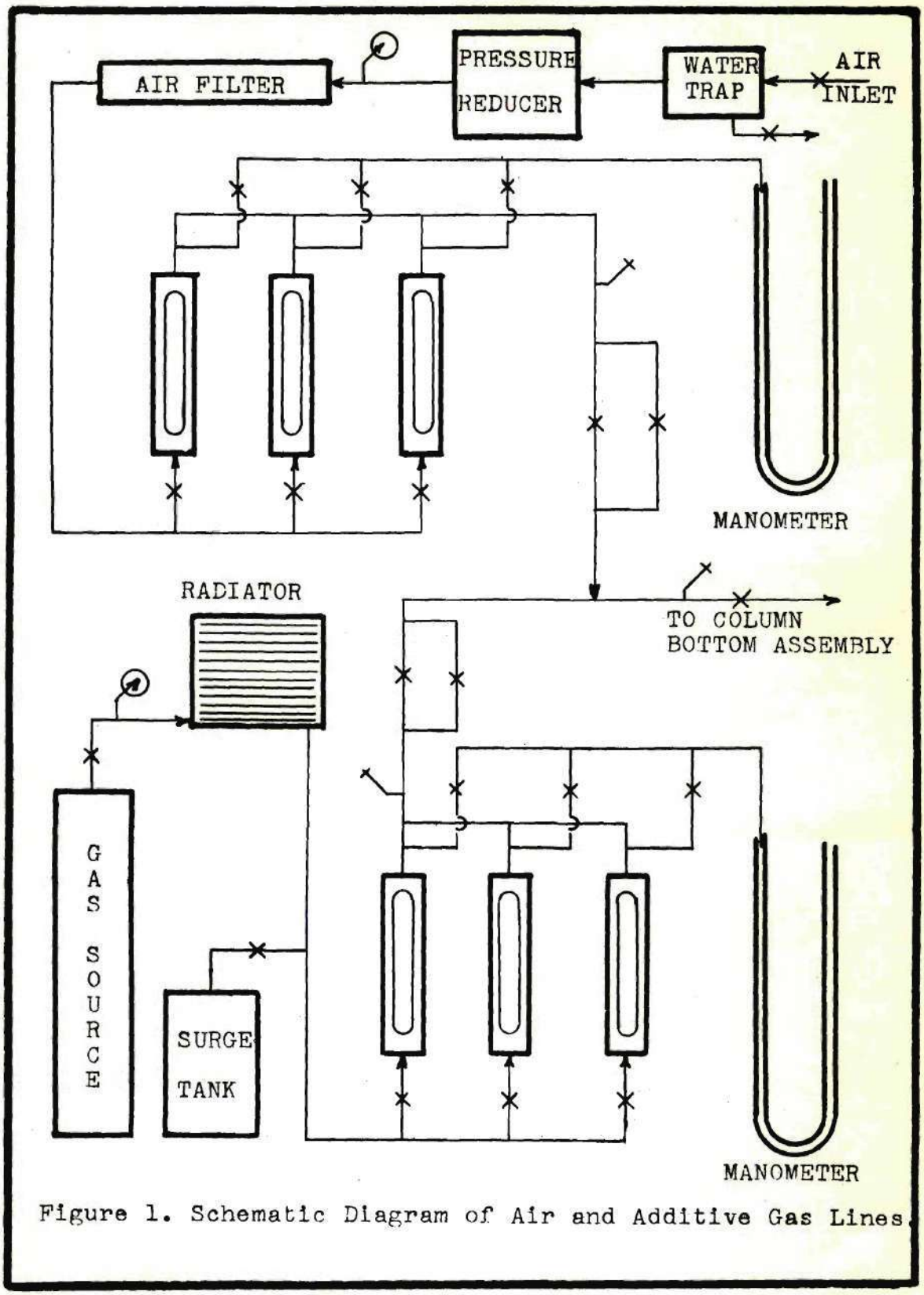


Figure 1. Schematic Diagram of Air and Additive Gas Lines.

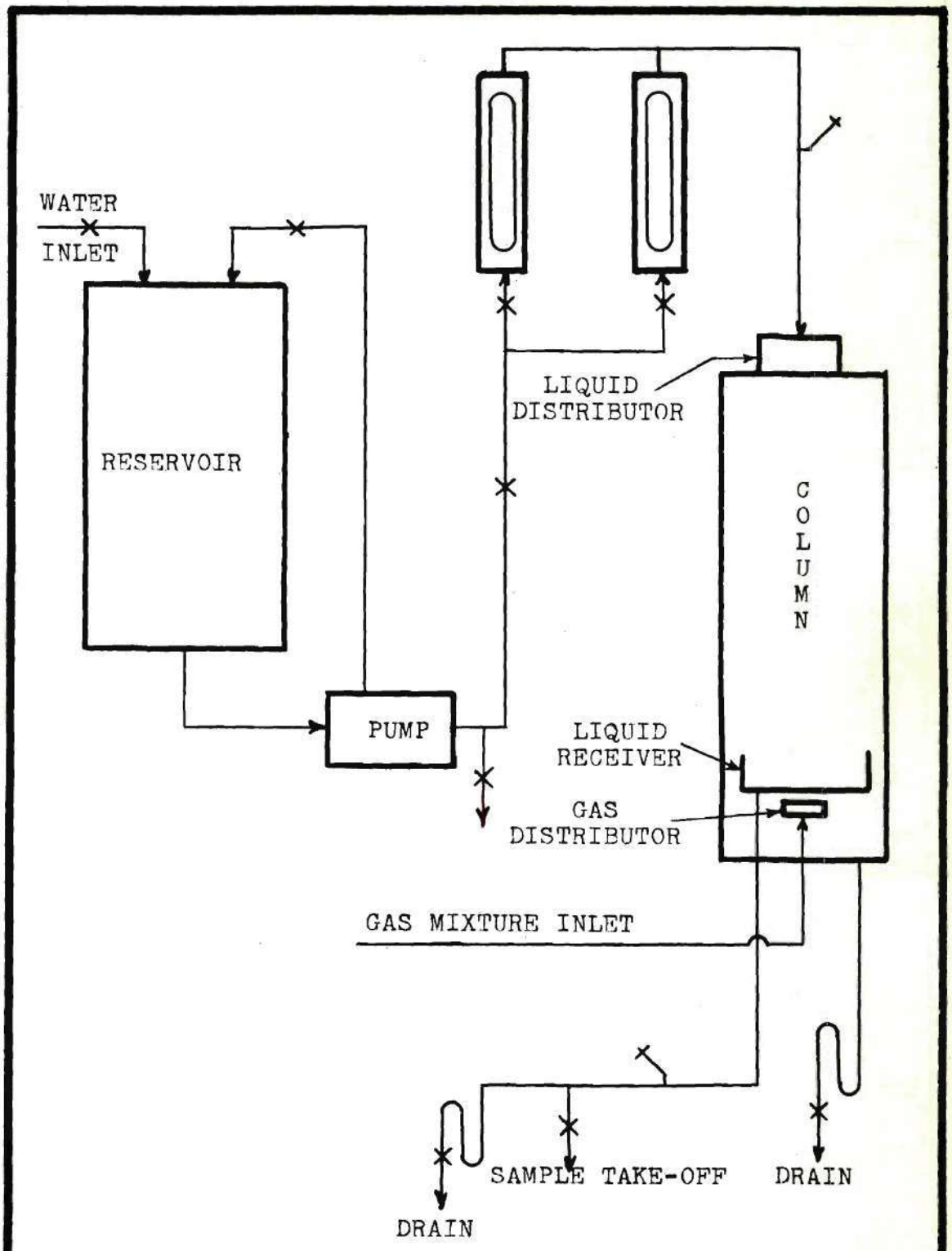


Figure 2. Schematic Diagram of Water System and Column.

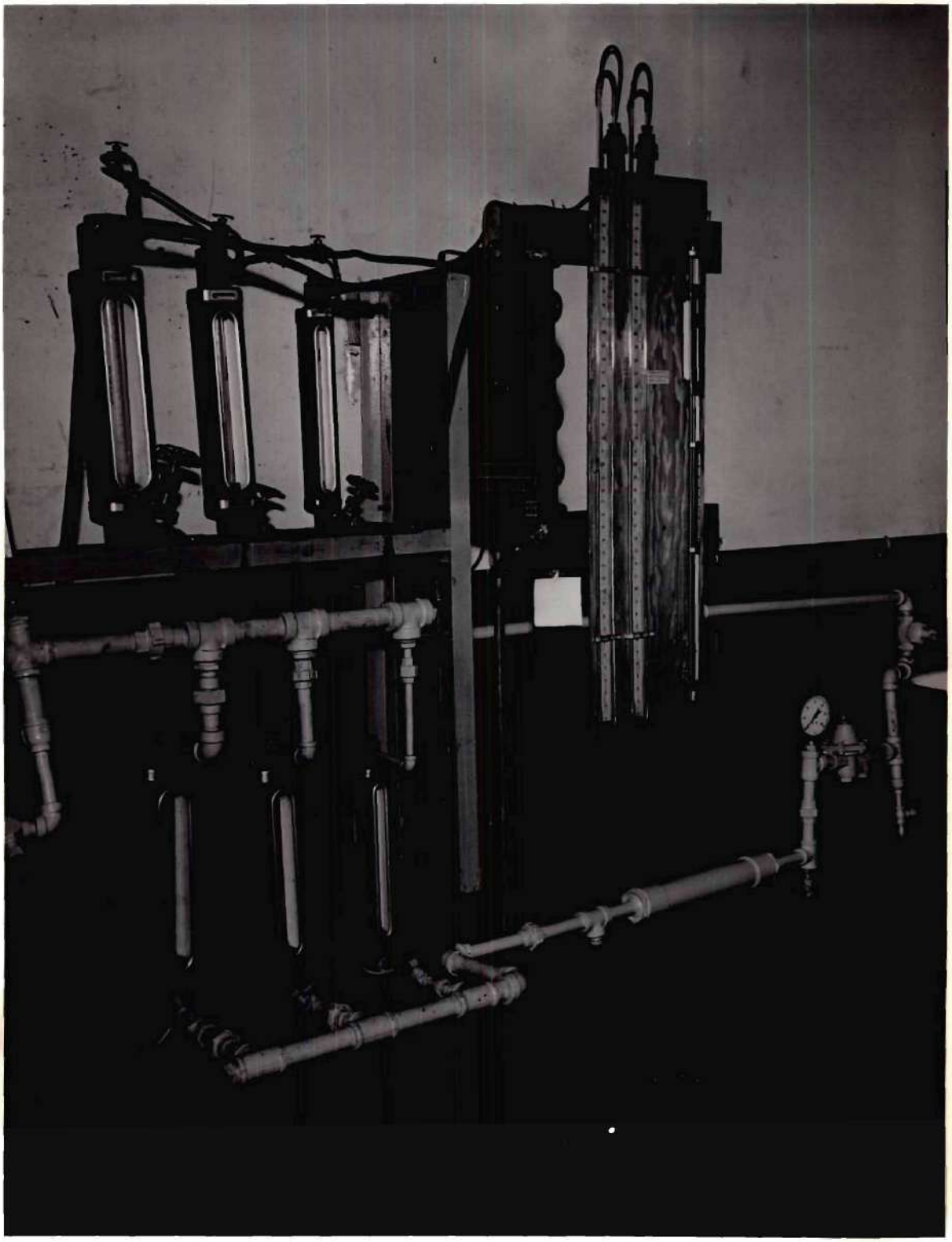


Figure 3. Photograph of Air and Gas Lines

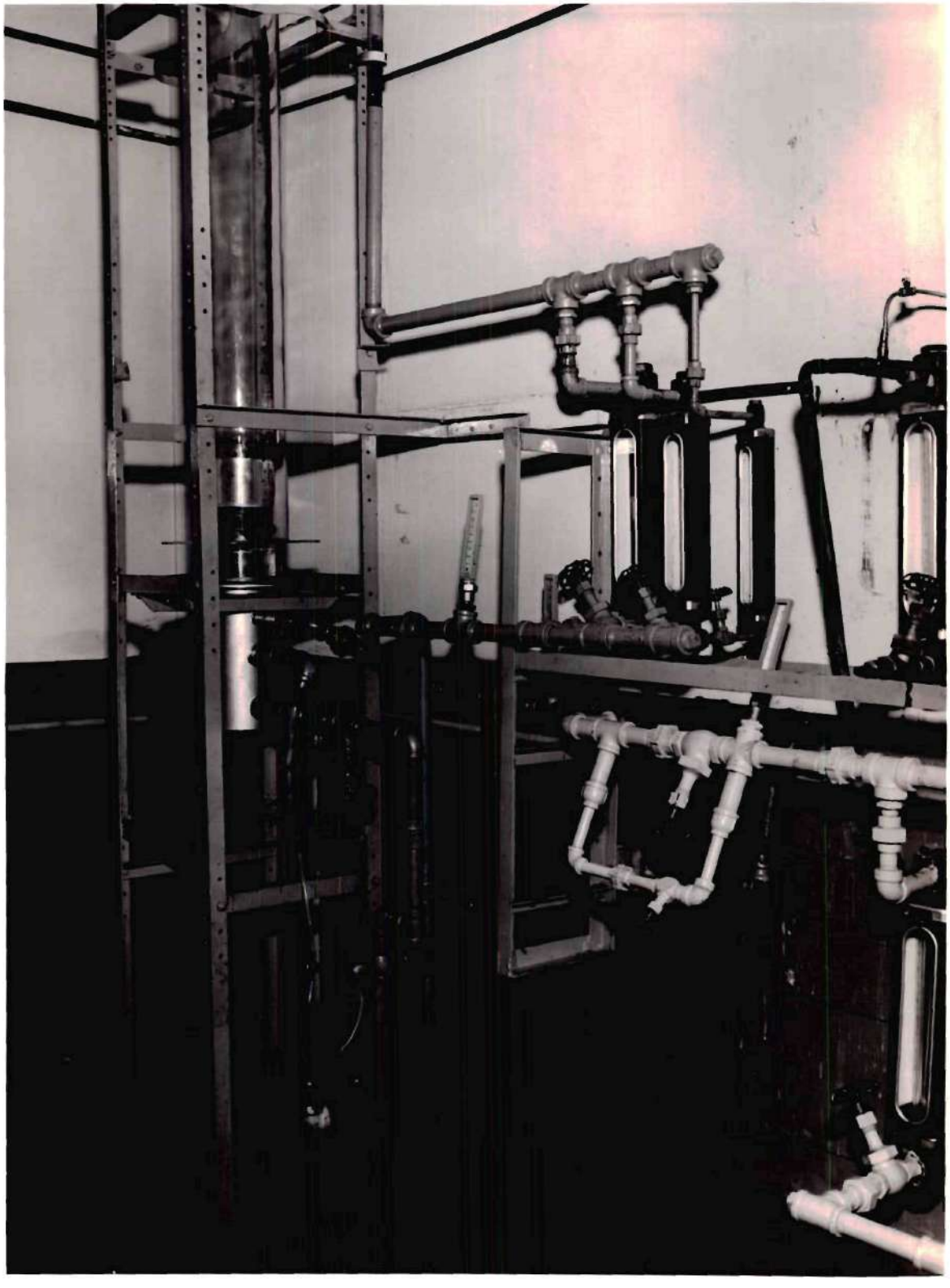


Figure 4. Photograph of Water System and Column Assembly

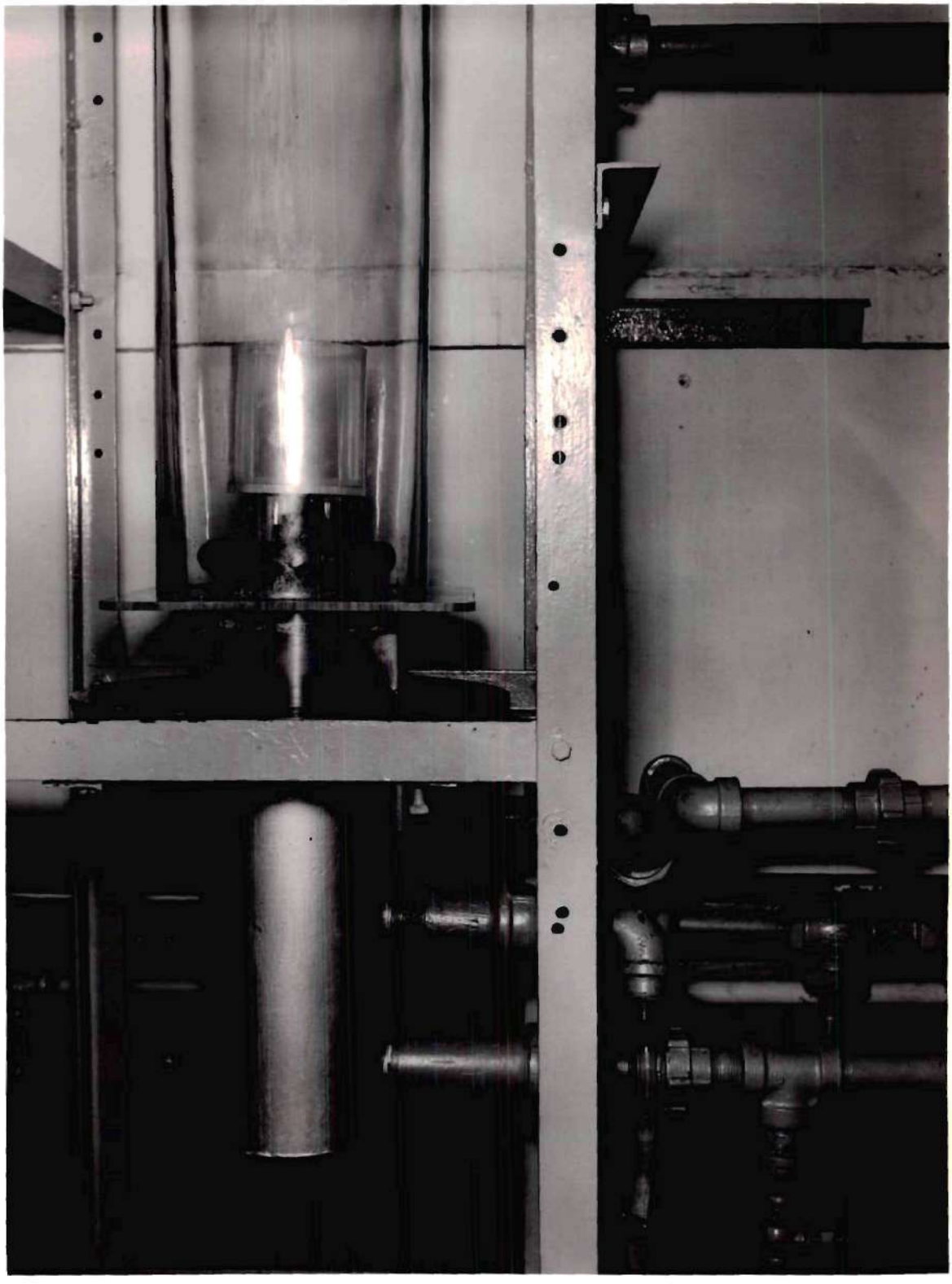


Figure 5. Photograph of Column Bottom Assembly

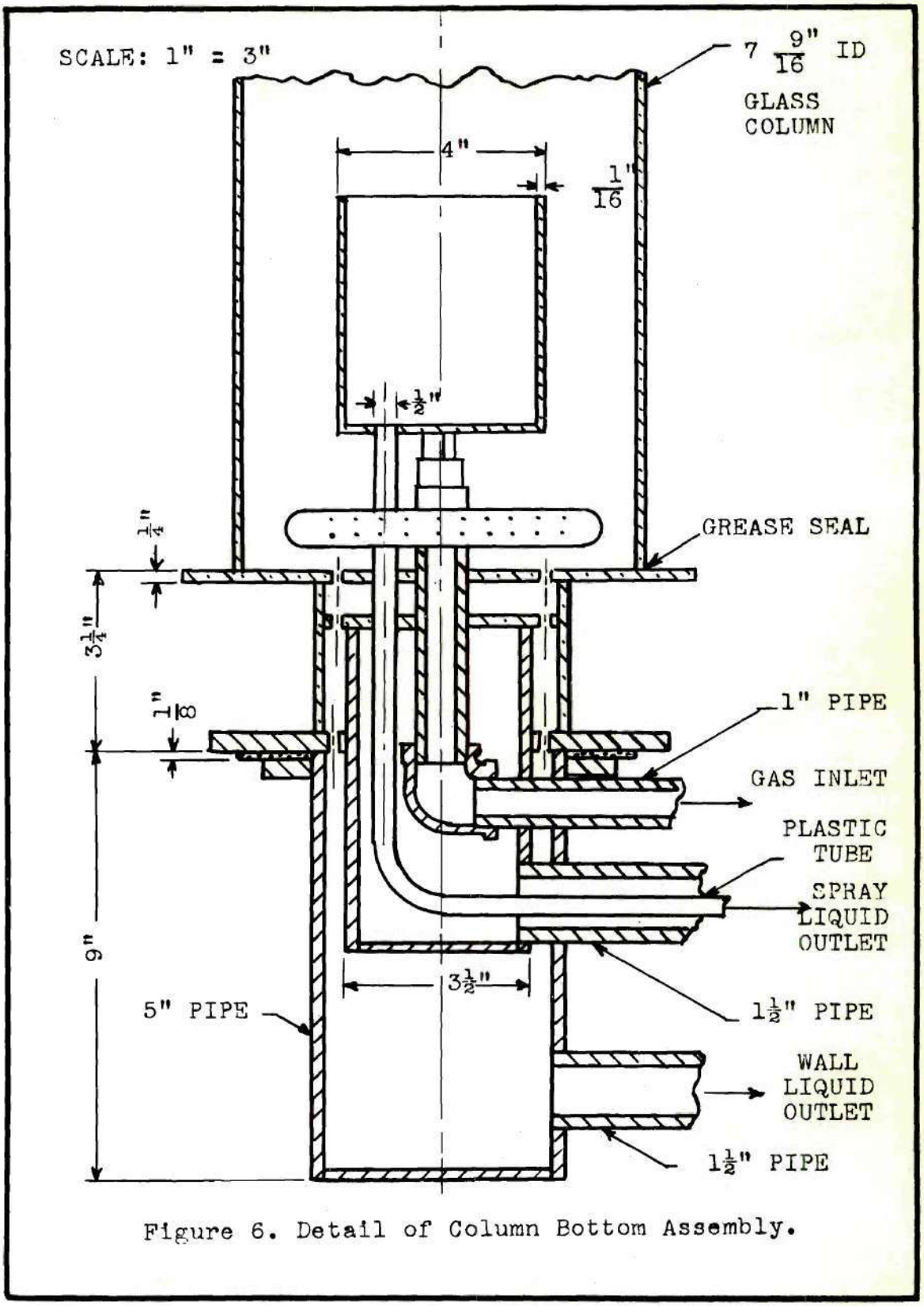


Figure 6. Detail of Column Bottom Assembly.

SCALE: 1" = 1"

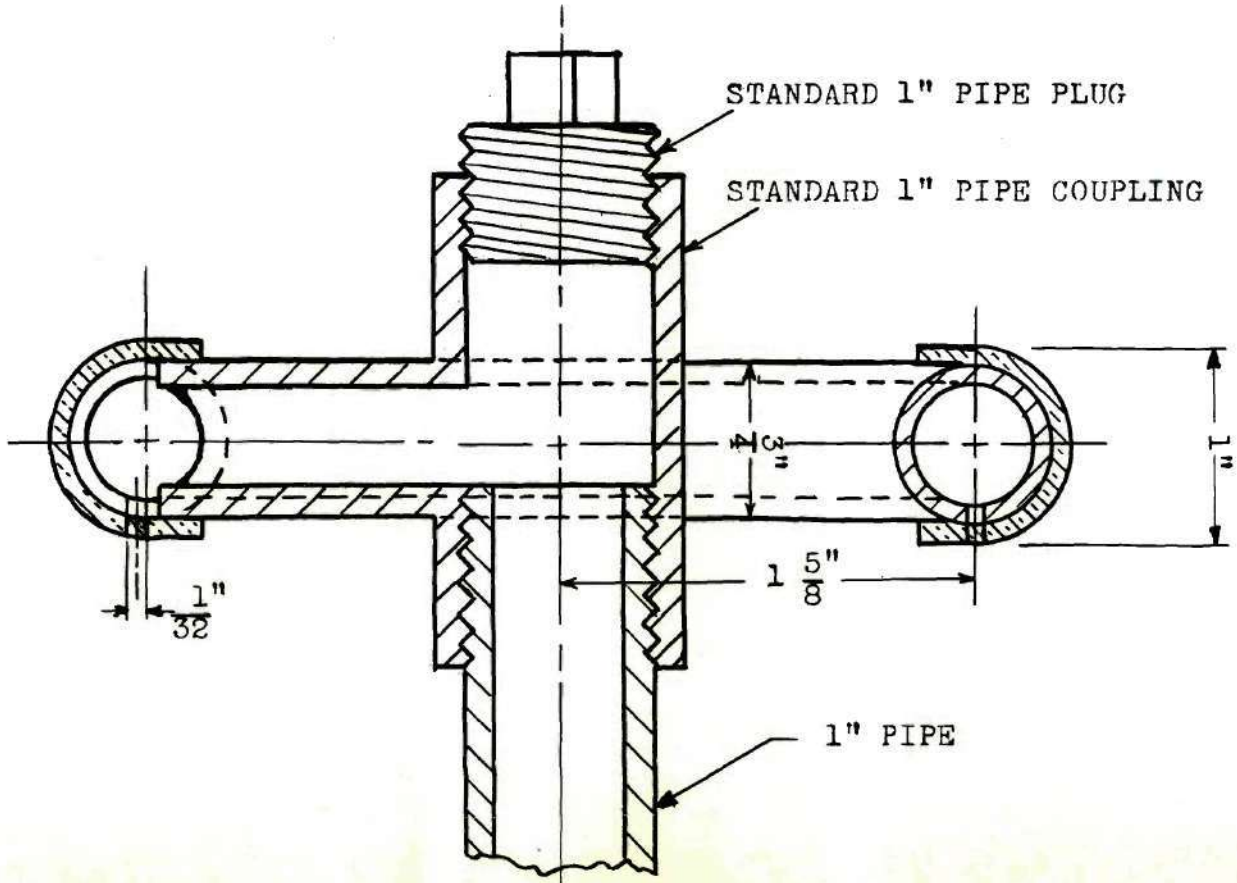
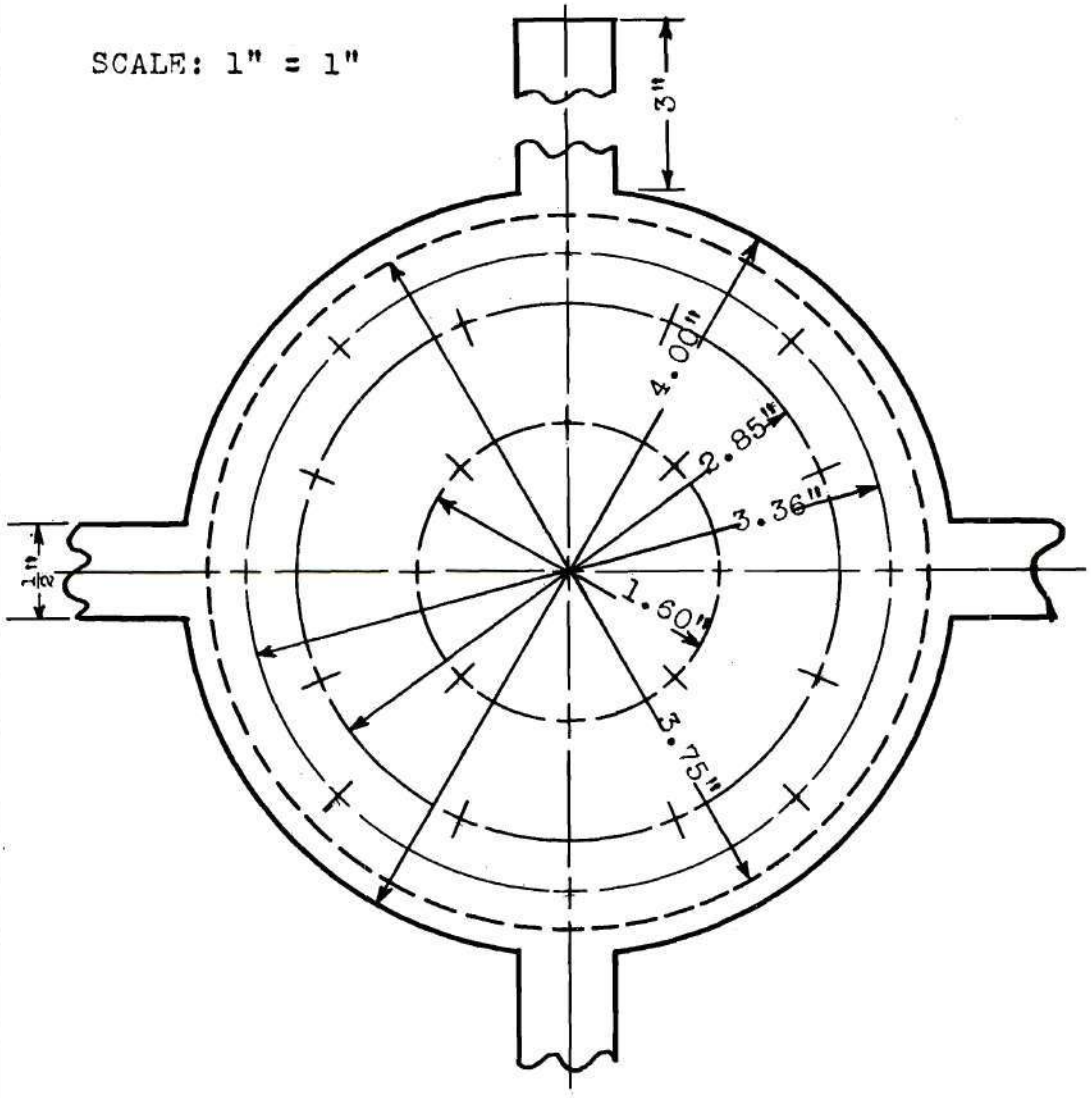
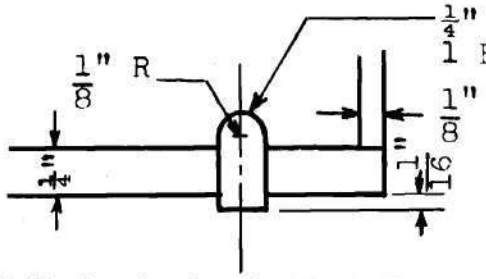


Figure 7. Detail Diagram of Gas Distributor.



SCHEMATIC LAYOUT OF DISTRIBUTOR HOLES



DETAIL OF DROPPER EMPLACEMENT

Figure 8. Diagram of Liquid Distributor.

CHAPTER IV

EXPERIMENTAL MATERIALS

Tap water, supplied by the Atlanta Water Works, Atlanta, Georgia, was used as the absorbing liquid for this investigation.

Carbon dioxide was obtained in cylinders from the Liquid Carbonic Corporation, of Atlanta, Georgia. The carbon dioxide was specified as 99.8 per cent or better carbon dioxide and approximately 0.2 per cent nitrogen.

Anhydrous ammonia was obtained in commercial cylinder form from Tesco Chemicals, Inc., of Atlanta, Georgia.

Sulfur dioxide, specified 99.99 per cent sulfur dioxide and 0.01 per cent water was supplied in cylinders by the Virginia Smelting Company, of West Norfolk, Virginia.

Compressed air was obtained from the central laboratory compressor.

Velocite Oil D, viscosity 100 seconds at 100°Fahrenheit was furnished by the Standard Oil distributor in Atlanta, Georgia. This was the oil used as a protective layer for the liquid contained in the drop receiver at the bottom of the column.

Surface active agents utilized were:

(1) Aersol OT, dioctyl-sodium sulfosuccinate, an anionic type surface active agent, made by the American Cyanamide Company.

(2) Sterox SK, chemically polyoxyethylene thioethers, that is, a condensation product of dodecyl mercaptans with ethylene oxide, a

non-ionic surface active agent, obtained from the Monsanto Chemical Company.

(3) Victawet 12, a non-ionic surface active agent, phosphorous base, medium length alkyl group, produced by the Victor Chemical Works of Chicago, Illinois.

Surface tension concentration data for these agents are tabulated in Table I, Appendix I, and are presented graphically in Figures 10, 11, and 12, Appendix II.

CHAPTER V

EXPERIMENTAL PROCEDURES

General procedure.--During the course of this investigation the same type of experimental data for all of the systems studied was encountered, therefore, a general procedure common to all of the experimental runs was established.

An experimental run was begun by filling the clean reservoir with tap water. If a surface active agent was used to lower the surface tension of the liquid, the amount of the agent needed to produce the desired concentration was added to the known amount of liquid in the calibrated reservoir. The centrifugal pump was then started and the liquid was recycled through the pump and back to the reservoir for a period of at least one half an hour to insure adequate mixing. Liquid samples were taken from the reservoir periodically and checked for uniformity of surface tension values using a duNouy Interfacial Tensiometer.

The additive gas was introduced to the system from a standard commercial cylinder. A pressure regulator at the cylinder outlet was used to reduce the pressure to the approximate operating range of ten psig. The desired additive gas flow rate was obtained at a rotameter pressure of ten psig by manual adjustment of the needle valve controlling the gas flow into the rotameter and the downstream needle valve governing the downstream pressure.

The air stream entered the system through a gate valve on the

central air supply line. The pressure was reduced to the approximate operating range of ten psig by a pre-set spring actuated pressure regulator. The desired air flow rate was then obtained at a rotameter pressure of ten psig by manual adjustment of the needle valve governing the air flow into the rotameter and a needle valve downstream controlling the rotameter pressure.

The resulting air-additive gas mixture was passed into the column. Before proceeding further, sufficient time was allowed to insure a complete flushing of the column by gas of the composition to be studied.

The water, with or without an added surface active agent as the case may be, was metered through a calibrated rotameter by manual operation of a needle control valve and then passed into the liquid distributor centered on the top of the column. The resulting drops from the liquid distributor were collected in the liquid receiver cup at the bottom of the column. The liquid in the receiver was covered with a one-half inch layer of Velocite Oil D to protect the collected liquid from further absorption of gas from the air-additive gas stream. The receiver was completely flushed of the absorbing solution several times after an equilibrium condition was reached to insure that a true representative liquid sample was obtained. The liquid level control valve on the receiver outlet line was set so as to maintain a constant premarked liquid level in the receiver.

Liquid samples were then removed from the sample take-off line below the liquid level control valve by calibrated pipette, placed in prepared sample flasks, and stoppered for analysis as described in Chapter VI, Analytical Methods. After the receiver liquid samples

were obtained, additional samples were removed from the top of the column, taken just as the drops left the machined brass drop rods. Known amounts were placed in sample flasks for analysis. Liquid samples were also periodically removed from the reservoir for analysis. Liquid samples from the top of the column were used to determine the amount of gas absorbed during the formation of the liquid drop, the bottom samples were used to obtain the amount of gas absorbed during the actual passage of the drop through the column, and the reservoir samples were utilized to check for the amount of standard solution used to neutralize the test liquid.

While the column was equilibrating, the number of drops per minute was determined for each hole by an actual visual timed count.

The temperatures of the air, additive gas, air-additive gas mixture, exit gas, entrance and exit water, and room were recorded along with the prevailing atmospheric pressure during the course of the experimental run. The air and additive gas flow rates and the liquid flow rates were recorded as were the results of the analysis of all of the liquid samples obtained.

After the data and liquid samples were obtained, the inlet water was turned off and the receiver drain line valve was closed. The next desired gas rate and composition was obtained through manual operation of the air and additive gas controls, and the entire procedure was repeated.

Comments on general procedure.---The column was frequently checked to insure that the aligned vertical position was maintained. All grease

seals were inspected periodically to insure against a gas leak at the bottom of the column.

The liquid distributor was marked and oriented so that the same position was maintained for all runs. The distributor was constantly checked during operation to make certain that none of the passages were blocked or obstructed by suspended particles in the liquid stream.

The gas distributor was checked occasionally to determine if the exit gas holes were clear and unobstructed.

The oil film on the surface of the liquid in the liquid receiver was constantly observed during the progress of an experimental run to insure that a properly consistent film was maintained.

After each series of runs involving one concentration of a surface active agent the entire system was completely flushed with water before introducing another mixture. Samples were taken of the next solution mixed in the reservoir and the surface tension measurements were compared to those obtained from liquid samples entering the liquid distributor.

The reservoir was completely flushed and scrubbed after use of each different agent was completed to remove all traces of the agent. The reservoir was also drained and cleaned at any other time when the cleanliness of the liquid solution was questionable.

When a different additive gas was introduced to the system, the entire additive gas and air-additive gas mixture lines were completely flushed over the period of one day.

CHAPTER VI

ANALYTICAL PROCEDURES

Carbon dioxide.--The liquid samples containing carbon dioxide in solution were removed at the liquid take-off point and at the column top in known amounts, and added to a known amount of a standard solution of barium hydroxide which had been previously placed in a sample flask. When the sample was added to the barium hydroxide solution a white precipitate of barium carbonate was formed. The resulting mixture was titrated with a standard solution of dilute hydrochloric acid to determine the excess of barium hydroxide present. The indicator used for the titration was phenolphthalein.

Sulfur dioxide.--The liquid samples of known volume were titrated with a standard solution of sodium hydroxide. The indicator used for the titrations was phenolphthalein.

Ammonia.--The liquid samples of known volumes were titrated with a standard solution of hydrochloric acid. The indicator used was phenolphthalein.

General practices.--The sample flasks were thoroughly cleaned after use, dried in an oven, and stoppered for further use. The standard solutions were checked before, during, and after the completion of the lengthy experimental runs to insure that the normality of the solutions were unchanged. The standard solution containers were equipped with calcium

chloride and ascarite absorption tubes to remove from the air entering the containers any water vapor and any of the gases used during the investigation which had not been exhausted from the room. The burette for the barium hydroxide was also equipped with a similar adsorption tube. All volumes were measured in precision burettes. All volumes of the liquid samples were measured in calibrated pipettes.

Surface tension.--The surface tension of the liquid was determined through use of a duNouy Tensiometer. This apparatus was calibrated with distilled water and with weights of known mass. The calibration of the Tensiometer was rechecked before each set of experimental determinations of the surface tension of the tested solutions.

CHAPTER VII

METHOD OF CALCULATION

An absorption column can be schematically represented by Figure 9 where G and L are the amounts of gas and liquid flowing through the column, respectively.

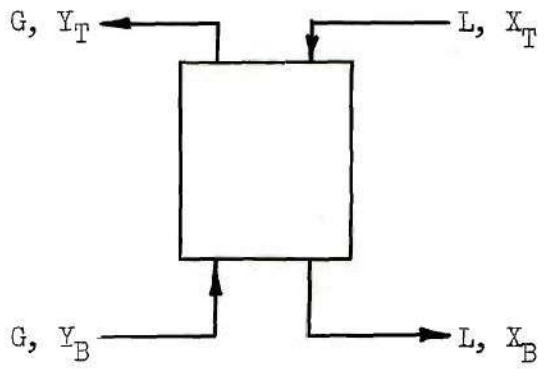


Figure 9. Schematic Diagram of an Absorption Column

Let X be the weight ratio of the solute to the solvent in the liquid stream, and Y be the weight ratio of the solute gas to the carrier gas in the gas stream. By a material balance,

$$L\,(X_B - X_T) = G\,(Y_B - Y_T) \tag{23}$$

where the subscripts B and T denote the bottom and top of the column, respectively.

If N_A is the amount of solute absorbed per unit area per unit time, a material balance covering a differential section of the column can be written as,

$$N_A a dV = K_L a(c_e - c)dV \tag{24}$$

where a is the effective area per unit volume.

V is the volume of the column.

K_L is the overall mass transfer coefficient.

c_e and c are the concentrations of the solute in equilibrium with the solute in the vapor phase and in the main liquid stream, respectively.

Since the amount of solute absorbed is the same as the material gained by the liquid phase or the material lost by the gaseous phase, the following equation can be written,

$$N_A = K_L(c_e - c) = L(X_B - X_T) \tag{25}$$

$(c_e - c)$ is actually the driving force which causes the solute to diffuse through the interface. Over an infinitesimal section of the column, equation (25) may be written as,

$$N_A = K_L(c_e - c) = LdX \tag{26}$$

Both X and c are used to express the amount of solute in the liquid phase; they may also both be expressed in terms of mole fraction, N. Consequently equation (26) may be expressed as,

$$N_A = K_L(N_e - N) = LdN \tag{27}$$

Over the entire length of the column, the overall transfer coefficient may be calculated as,

$$K_L = \frac{LdN}{N_e - N} \tag{28}$$

The driving force, now expressed as $(N_e - N)$ changes in the column as the concentration of the solute in the liquid stream changes. In the case where Henry's law can be applied, both the operating and the equilibrium line may be considered straight over the range investigated, and the logarithmic mean of the driving force may be used to calculate the overall coefficient. Thus equation (28) may by expressed as,

$$K_L = \frac{N_A}{(N_e - N)_{lm}} = \frac{N_A}{\frac{\Delta N_T - \Delta N_B}{\ln\left(\frac{\Delta N_T}{\Delta N_B}\right)}} \qquad (29)$$

where $\Delta N = (N_e - N)$.

The average diameter of the liquid drops, D_D in inches was calculated through the use of the experimentally determined average number of drops per individual hole per minute, N_D, in the following manner.

The number of drops per hole per minute times the total number of holes, N_H, would give the total number of drops per minute as,

$$N_D N_H = \text{total drops per minute} \qquad (30)$$

The total liquid flow rate, L in pounds of water per minute, divided by the total number of drops per minute would yield the pounds of water per drop, expressed by,

$$\frac{L}{N_D N_H} = \text{pounds of water per drop} \qquad (31)$$

Assuming that each drop is a perfect sphere of diameter D_D in inches, the volume per drop expressed in cubic feet would be given by,

$$\frac{\pi}{6}\left(\frac{D_D}{12}\right)^3 = \text{cubic feet of water per drop} \qquad (32)$$

The pounds of water per drop may then be obtained by multiplying equation (32) by the density of water at the experimental conditions, as follows,

$$\frac{\pi}{6}\left(\frac{D_D}{12}\right)^3 \rho\, S_g = \text{pounds of water per drop} \qquad (33)$$

where ρ is the density of water in pounds per cubic foot at 32° Fahrenheit and S_g is the specific gravity of water at experimental conditions based on 32° Fahrenheit.

Equating equations (31) and (33) and solving for D_D, we obtain,

$$D_D = 12\left(\frac{6L}{N_D N_H \pi \rho\, S_g}\right)^{1/3} \qquad (34)$$

The total area available for absorption, A in square feet is calculated using the expression for the average drop diameter, D_D from equation (34) and the number of drops per hole per minute, N_D.

The total surface area, in square feet per drop, assuming a perfect sphere may be expressed as,

$$\pi\left(\frac{D_D}{12}\right)^2 = \text{square feet of surface area per drop} \qquad (35)$$

The total surface area per drop multiplied by the number of drops per hole per minute and the number of holes would give an expression for the square feet of surface area presented in the column per minute, as

$$\pi\left(\frac{D_D}{12}\right)^2 \cdot N_D N_H = \text{surface area in square feet per minute} \qquad (36)$$

The time of fall for the drop may be computed by

$$S = 1/2\ gt^2 \qquad (37)$$

where S is the total displacement in feet.

g is the local acceleration due to gravity in feet per second^2.

t is the time in seconds.

The length of fall was constant over the course of the entire investigation, consequently, the time of fall or resulting time of exposure for the drop was constant and calculated as 0.007917 minutes.

The total area presented for absorption may then be expressed as,

$$\text{Total area in square feet} = \pi\left(\frac{D_D}{12}\right)^2 N_H N_D\ (0.007917) \qquad (38)$$

Substituting for D_D from equation (34), and clearing of known constants, we obtain finally,

$$\text{Total area in square feet} = 4.47 \times 10^{-3}\left(\frac{N_D}{S_g^2}\right)^{1/3} \qquad (39)$$

CHAPTER VIII

RESULTS AND DISCUSSION

In this investigation the rate of absorption of carbon dioxide, sulfur dioxide, and ammonia by liquid drops was studied in a seven and nine-sixteenth inch inside diameter spray type column using tap water as the absorbent and varying the surface tension of the liquid from 72.4 to 25.2 dynes per centimeter through the use of three surface active agents, Aersol OT, Sterox SK, and Victawet 12. A complete summary of the results may be found in Tables II, III, and IV of the Appendix. Due to the large number of experimental runs only portions of the data will be shown in graphic form.

One liquid rate of 0.5 pounds of water per minute was employed throughout the entire investigation, consequently, the effect of variable liquid rates on the rate of absorption of the various gases was not determined. Gas rates ranging from 0.0088 to 0.143 pounds per minute were used in the study of carbon dioxide with the concentration of the diffusing gas in the entering gas stream being maintained at 100, 80, or 65 per cent by volume. The range of gas flow rates for sulfur dioxide was from 0.0145 to 0.200 pounds per minute and sulfur dioxide concentrations of 50, 30, and 10 per cent by volume were employed. In the ammonia study, gas rates from 0.281 to 1.690 pounds per minute were used with ammonia concentrations in the entering gas stream maintained at 5, 3, or 1 per cent by volume.

Since the conditions at the gas-liquid interface were not known, the conventional practice of reporting results as over-all transfer coefficients instead of individual film coefficients was followed in this work. The calculations involving the determination of the over-all coefficients from experimental data are described in Chapter VII, Methods of Calculation. These coefficients for carbon dioxide and sulfur dioxide were corrected in each case to a base temperature for comparison purposes by the method of Sherwood and Holloway (8).

A. Absorption of Carbon Dioxide

1. Effect of gas rate.--Results of the study of the absorption of carbon dioxide by drops of plain tap water in the spray type column indicate that the over-all mass transfer coefficient for the liquid film is not affected by gas flow rates. Since carbon dioxide is only slightly soluble in water, most of the resistance to mass transfer should be in the liquid film which itself is relatively independent of the gas stream conditions. The experimental data, therefore, concurs with the results expected. Selected data are presented in Figure 13 of the Appendix.

It should be noted that the gas velocities encountered in the course of the study of carbon dioxide absorption were in the range from 0.25 to 4.50 feet per minute so that the Reynold's numbers varied from 120 to 469. Thus, even if the over-all transfer coefficient were affected to some limited extent by gas velocity, it is doubtful if this effect could be observed over the range of gas velocities investigated.

The amount of carbon dioxide absorbed by the liquid drops during the period of formation on the liquid distributor was negligible. This may be attributed to the slight solubility of carbon dioxide in water

in addition to the short time, from 0.3 to 0.6 seconds, that the drop was exposed on the distributor tap. It should be noted that during this short period of time, the drop must essentially cover the range from a condition of no surface area, i.e., a condition in which there is only a liquid film of negligible thickness on the liquid distributor tap immediately after the drop has fallen off, to a full formed drop just leaving the tap. A final factor for consideration is the obstruction of the column exit by the liquid distributor. Due to the low gas velocities encountered during the carbon dioxide study, the flow pattern of the gas in the column was one of streamline motion which by-passed the column exit obstruction made by the liquid distributor. This topic will be more fully developed later.

2. Effect of concentration of carbon dioxide.--The results of the study of the effect of the concentration of carbon dioxide in the gas stream upon the rate of absorption of carbon dioxide indicate that this variable had no noticeable influence on the over-all coefficient. This is in agreement with gas absorption theory since an increase in the concentration of the diffusing gas in the main gas stream will increase the driving force which will in turn increase the total moles absorbed per unit time per unit area. These two effects should tend to offset each other in the final calculation of the over-all mass transfer coefficient. A graphical presentation of selected data appears in Figure 14 of the Appendix.

3. Effect of surface active agents.--The results of the study of carbon dioxide on the absorption of carbon dioxide indicate that there is no effect on the over-all mass transfer coefficient exerted by the presence

of surface active agents in various concentrations in the liquid phase. This is illustrated by Figures 15 and 16 of the Appendix.

The observed effect was that the moles of carbon dioxide absorbed per unit of time increased with a decrease in surface tension. However, this increase was accompanied by a corresponding increase in the area available for absorption due to the fact that the number of drops per unit volume of the liquid increased. The relationship between the number of drops per unit volume of entering liquid and the area available for absorption is given in Chapter VII, Methods of Calculation. The value of the logarithmic mean driving force was influenced only slightly by the increase in the absorption of carbon dioxide. Consequently in the calculation of the over-all coefficient, the increase in the moles absorbed per unit time and the available area for mass transfer counteracted each other and the over-all coefficient was thus unaffected.

B. Absorption of Sulfur Dioxide

1. Effect of gas rate.---A comparison of the over-all transfer coefficients obtained for the absorption of sulfur dioxide at various gas flow rates indicated that the gas velocity had no effect on the over-all coefficient. This is in agreement with the expected result since the resistance of the liquid film is the controlling factor, and the results should parallel those for carbon dioxide absorption. Data illustrating these results is presented in Figure 17 of the Appendix.

It should be noted that the gas velocities are so low, from 0.40 to 5.00 feet per minute, that even if the over-all mass transfer coefficient were affected to some limited extent by gas velocity, it is doubtful if this effect could have been observed during this investigation.

The amount of sulfur dioxide absorbed by the liquid drops during the period of drop formation was in the range of two to three per cent of the total amount of sulfur dioxide absorbed during the total period of formation and fall. This increase over that of the carbon dioxide study is attributed to the fact that sulfur dioxide is more soluble in water than is carbon dioxide.

2. Effect of concentration of sulfur dioxide.--The results of this study indicate that there was no influence of the concentration of the diffusing gas in the main gas stream on the value of the over-all absorption coefficient. The explanation of these results is based on the same assumptions as were presented for the carbon dioxide study since both gases are only slightly soluble in water and, therefore, should exhibit similar properties in absorption studies. Selected data is presented graphically in Figure 18 of the Appendix.

3. Effect of surface active agents.--No effect of surface active agents on the over-all absorption coefficient for sulfur dioxide was observed. The results are in agreement with those observed and presented for carbon dioxide. Figures 19 and 20 of the Appendix illustrate this point.

C. Absorption of Ammonia

1. Effect of gas rates.--The results of this study indicated that the over-all mass transfer coefficient of absorption for ammonia increased with increasing gas velocity or Reynold's number of the gas phase. A plot of the logarithm of the over-all absorption coefficient versus the logarithm of the Reynold's number of the gas stream yielded a correlation of the form,

$$K_G = a(Re)^b$$

where: K_G is the over-all mass transfer coefficient in terms of pound moles per hour-square foot-atmosphere.

a is a constant

b is a constant

Re is $Du\rho/\mu$, the Reynold's number of the gas stream.

Substitution of experimentally determined values gives,

$$K_G = 0.213\ Re^{0.25}$$

Graphical presentation of this correlation appears in Figure 21 of the Appendix.

This is in agreement with the expected results, since the absorption of ammonia constitutes a case in which the gas film is the controlling factor. Any change in the condition of the gas film would then exert some influence upon the over-all coefficient. In this case, an increase in the gas velocity tends to diminish the thickness of the film and thus decrease the resistance of the gas film to mass transfer.

The amount of ammonia absorbed during the period of drop formation varied from thirty to fifty per cent of the total amount of ammonia absorbed. The amount of ammonia absorbed by the drop during the formation period decreased with a decrease in the gas flow rate. Thus, this effect was attributed to a change in the flow pattern of the gas stream near the exit of the column. Relatively high gas rates, from 12.63 to 75.68 feet per minute, forced the main gas stream to hit directly on the liquid distributor at the top of the column, and as the flow rate decreased there was less turbulence in the gas stream around the obstruction.

However, the greater portion of the increase in the absorption of ammonia during the period of drop formation over that found in the carbon dioxide study was due to the higher solubility of ammonia in water.

2. Effect of concentration of ammonia.--The results of this study indicate that the concentration of ammonia in the main gas stream had no effect on the over-all mass transfer coefficient. The explanation of these results is based on the same assumptions previously presented in the carbon dioxide discussion. Selected data is shown in Figure 21 of the Appendix.

3. Effect of surface active agents.--The over-all coefficient was found to be independent of the concentration of the surface active agents in the liquid phase over the range studied. The effect of surface tension upon gas absorption has been previously discussed for carbon dioxide, and the same reasoning also applies to ammonia. Selected data is shown in Figures 22 and 23 of the Appendix.

A comparison of the experimental values obtained for K_L and K_G during this study with those of other investigators is not possible since there are no values reported in the literature for this type of investigation.

This concludes the presentation of the experimental results of this investigation, as well as the discussion of the influence of the gas flow rate and the concentration of the diffusing component in the gas stream on the over-all mass transfer coefficient. However, there remains to be made some comments regarding the effect of surface tension and the effect of the concentration of the agent used to lower the

surface tension upon the over-all mass transfer coefficient.

D. Surface Tension and Surface Active Agents

The effect of surface active agents and surface tension upon the rate of mass transfer has not been investigated very extensively as the small number of references shows. However, even so, this small number of studies have reported a wide variety of results. These results range from an observed increase in the rate of absorption to a reported decrease in the rate of absorption when surface active agents are used to alter the surface tension of the absorbing liquid.

A basic point involving film structure should be clarified before proceeding further in the discussion of surfaces and surface films. Various investigators (27,28,29,30) have shown that the rate of evaporation of liquids is decreased when a mono-molecular layer film of various immiscible oils is present upon the liquid surface. This is undoubtedly a correct result, however, it does not have any necessary connection either to systems to which surface active agents have been added or to the surface concentration of such added surface active agents as determined by the Gibbs equation. The condition of a mono-molecular layer of oil upon the liquid surface actually represents two immiscible liquids, the oil being less dense and, consequently, settling upon the top of the heavier water. Under these conditions there is no reason why the rate of evaporation of the liquid should not show a noticeable decrease since the effective area for mass transfer has been covered by the oil layer and the liquid molecules must now move through the covering oil layer by diffusion to reach the gas phase. This is in direct contrast with the structure of the Gibbs surface. In this case the surface active agent

molecules are actually a part of the solution and thus not an immiscible film floating on another liquid phase. Now the molecules in the Gibbs surface are free to move into the liquid bulk, and the surface may be visualized as in a state of continual motion. It is most important that the basic difference between an oil layer upon the liquid surface and a Gibbs theoretical surface be noted.

When a Gibbs surface is assumed, calculation of the excess surface concentration of the surface active agent on the liquid surface may be made. The value of the excess surface concentration has been determined for each of the surface active agents used in this investigation over the range of concentration investigated to establish the form of the bulk concentration versus excess surface concentration curve. This data is presented in tabulated form in Table I of the Appendix and is illustrated graphically in Figures 10, 11, and 12 of the Appendix. It should be noted that the bulk concentration-excess surface concentration curve does not follow a general pattern for all surface active agents, but varies for each individual agent since each exhibits its own particular surface tension-concentration characteristics and the slope of the surface tension versus concentration curve is the primary factor which determines the bulk concentration-excess surface concentration curve.

In the low concentration range as the concentration of the surface active agent in the bulk of the liquid approaches zero, the slope of the surface tension-concentration curve may attain any value. However, a consideration of the basic properties of the excess surface concentration shows that if there is no surface active agent in the solution, there can be no excess surface concentration. Thus, the value of the excess

surface concentration must approach zero as the concentration of the surface active agent in the liquid bulk approaches zero. This will apply to all surface active agents regardless of the characteristics of the surface tension-concentration curve.

In the higher concentration range, that range of concentration where the addition of more of the surface active agent produces little or no change in the value of the surface tension of the solution, the value of the excess surface concentration will again approach zero. Although the concentration of the agent in the liquid bulk is a sizable amount, the slope of the surface tension-concentration curve in this range is a small value and is approaching zero. Therefore, since the excess surface concentration is the product of concentration and slope, the excess surface concentration will also approach zero. In fact, if no change is noted in the surface tension of the solution upon an increase in the agent concentration, the slope of the surface tension-concentration curve is zero, and the value of the excess surface concentration will also be zero. Thus, the excess surface concentration at the limits of the surface active agent concentrations investigated in this study have been defined. Any variations in the value of the excess surface concentration must now appear in the rather small concentration range between zero and that condition of more or less constant surface tension. Reference to Figure 10 of the Appendix serves to illustrate this situation for the case of Aersol OT. It is seen that the value of the excess surface concentration reaches a maximum in the concentration interval stated above, and this is true for all surface active agents which lower the surface tension. The location of this maximum value of the excess surface

concentration depends upon the characteristics of the surface tension-concentration curve. More than one maximum may be noted, for example, a surface active agent which lowers the surface tension of the solution as the concentration of the agent in the bulk is increased, then exhibits an increase in surface tension with an increase in concentration, and finally again exhibiting a decrease of the surface tension with an increase in concentration, as is the case of $K_2Cu(CN)_3$. Further discussion of effects of this type are beyond the scope of this investigation. It is most important to note that the wide variation in the value of the excess surface concentration from zero to a maximum value and thence again to zero occurs at very low concentrations of the surface active agent. A large variation in the surface tension of the solution, from 72.4 dynes to a low nearly constant value, also occurs in this same concentration range.

Consideration must be given to the structure of the agent molecule itself. It would not be possible to determine exactly the amount of the total surface covered by a surface active agent unless information were available regarding the orientation of the molecules at the liquid surface, the effective area covered per molecule, and the structure of the molecule.

It must be noted that although the Gibbs surface may exist, it is but one molecule thick. Therefore, even if this surface layer exhibited a ten or twenty fold increase in resistance to mass transfer, upon the addition of surface active agents the effect on the over-all mass transfer coefficient would be negligible when compared to the resistance and the relatively large thickness of the controlling liquid film. If the

Gibbs surface itself were able to exert a sizable influence on the rate of mass transfer then the rate of absorption would be expected to decrease due to either the increased resistance to diffusion of the liquid film or the actual effective blocking or covering of the available area for mass transfer by the surface active agent molecules in the Gibbs surface or a combination of these two effects.

Consideration of the case of the absorption of gases by a smooth liquid surface by Becker (2) and Hutchings (4) in a batch process led them to advance the theory that the rate of absorption was increased due to a decrease in the resistance to mass transfer in the liquid film caused by mechanical mixing. However, this investigator is of the opinion that turbulence is only one of the factors which influence the rate of absorption; the other is the increase in effective area through the continual presentation of a new surface for absorption and due to the added effects of splashing, ripple formation, and similar surface effects. It is, however, more or less a matter of a point of view as to whether or not this increase in absorption with stirring is due to an increase in the effective area for mass transfer or a decrease in the resistance of the liquid film.

The studies of Hedestrand (51) and Harkins (52,54) on the effect of a Gibbs surface on the rate of mass transfer show that no change in the rate of evaporation is observed. These results led Harkins to advance the theory that the Gibbs surface is not a static condition as is the case when a mono-molecular layer of oil is present upon the surface of the liquid. The manufacturers of Aersol OT (53) specify also that this material does not alter the rate of evaporation from a liquid surface.

Consider now the conditions existing in a wetted wall column.

Trenovoskaya and Belopolakii (39) observed a decrease in the absorption of sulfur dioxide in a wetted wall column when various surface active agents were employed. They attributed this phenomena to the presence of large amounts of the agent molecules upon the liquid surface as predicted by the Gibbs equation. They noted that a minimum absorption rate was attained at that agent concentration which showed a maximum value for the Gibbs excess surface concentration. It is most significant that they also reported a change in the flow pattern of the wall liquid, in this same concentration range. This has also been noted by Yu (76) and Engel (77). The change in the flow pattern of the wall liquid was observed at very low concentrations of surface active agents, in the same range of concentration where the maximum in the value of the excess surface concentration is also found. The flow pattern observed with plain water in a wetted wall column is one of a series on concentric ripples, however, with a decrease of the surface tension of the liquid caused by the addition of surface active agents, the concentric ring pattern disappeared, and it became difficult to visually detect the presence of liquid flow of the wall film. It is postulated by this investigator that the effective area for mass transfer may be decreased by decreasing the surface tension of the liquid which eliminates the concentric ripple pattern. The amount of surface area reduction depends upon the size, shape, and frequency of the concentric ripples. In addition it should be noted that the presence of ripples in the flow pattern of the wall liquid results in additional turbulence in the liquid film. This turbulence, however, decreases with the decrease in the

concentric ripple pattern thus affecting an increase in the resistance of the liquid film.

If the Gibbs surface is used to explain the reported decrease in absorption, one may doubt as to whether there are sufficient agent molecules available in the thin wall liquid film to provide the Gibbs excess surface concentration without seriously altering the liquid bulk concentration. This point is clarified in Appendix III where it is shown that there will always be sufficient molecules in the bulk liquid and the amount of agent removed from the liquid bulk by the Gibbs surface film is negligible. However, it should be noted that if Trenovoskaya and Belopolokii (39) attempt to explain their results through the use of the Gibbs surface phenomena, the rate of absorption should increase from the minimum value attained and approach the rate of absorption reported for plain water as the value of the Gibbs excess surface concentration tends to zero in the portions of the surface tension-concentration curve where little or no change in the surface tension of the liquid is observed with an increase in the concentration of the surface active agent. This result was not found. This investigator thus postulates that the decrease in the rate of absorption of sulfur dioxide observed by the above workers was due entirely to a decrease in the available surface for absorption through a change in the wall liquid flow pattern and a decrease in the turbulence of the liquid film due to the resulting disappearance of the concentric ring flow pattern.

It should also be noted that if the Gibbs surface concentration is used to explain the decrease in absorption, when the more or less constant value of the surface tension of the solution is obtained the

value of the Gibbs excess surface concentration has been shown to approach zero. In this case we would then predict that the rate of absorption should increase to attain approximately the same rate as observed with plain water. This, however, is not the case; a minimum absorption rate is obtained at a low concentration of the surface agents and is unaffected by a further increase in the concentration of the agent in the liquid bulk.

Packed columns do not afford conditions suitable for a comprehensive study of the effect of surface tension and surface active agents upon absorption. The work of Riou (24) serves to illustrate this point. In a study of the effect of surface active agents upon the absorption of carbon dioxide in a packed column, he obtained results ranging from an increase in the rate of absorption to a decrease in this rate. However, with the combination of the absorption coefficient and the area per unit volume into one experimental determined variable, we are unable to explain the significance of these results since we know neither the effect upon the liquid film coefficient nor the effect upon the area factor. Killfer (25) restudied the systems utilized by Riou (24) and concluded that the rate of absorption is increased and that this is due to a decrease in the surface tension of the liquid. The result appears acceptable since the area available for absorption would be expected to increase with a decrease in the surface tension of the liquid due to the increased wetting of the packing. This result, however, should not be thought of as a general one since it would be expected to vary with the characteristics of the agent employed, i.e., wetting ability, foaming tendency, etc. Thus it would appear that any effect upon the rate

of absorption could be accounted for in one manner or another.

It has already been shown that the presence of a Gibbs surface did not influence the rate of evaporation of liquids from a flat quiescent surface and, therefore, even if the formation of a Gibbs surface were plausible under the turbulent liquid flow conditions present in a packed column, no effect would be expected to be exerted upon the rate of absorption.

It is thus seen that packed columns do not afford conditions favorable for a study of the effect of surface tension and surface active agents upon absorption due to the direct combination of the two most important variables into one experimentally determined inseparable variable. It is also noted that this variable will be affected by certain characteristics of each surface active agent and the type of packing utilized.

The results of studies in spray columns are not easily evaluated. It was the original intent of this investigator to study the effect of surface active agents upon the absorption of various gases in a spray column utilizing a standard commercial type spray nozzle. The column bottom assembly was designed and constructed to permit a separation of the spray core from the wall liquid in such a column. The idea was to analyze both the spray and the wall liquid to determine the effect of surface active agents on the absorption of gases by each. The wall liquid was found to be in an unsteady state flow condition due to the large amount of turbulence in the column as a result of the spraying of the water into the column under pressure. The amount of wall liquid varied with the spray pattern due to the variance of the spray angle. The

wall liquid distribution thus had to be determined for each set of operating conditions. The spray nozzle delivered a different spray pattern with increasing flow rate and, consequently, the effective surface area was changed. This would have meant an evaluation of the drop size distribution for the spray at each liquid rate, however, this was in turn complicated by the added effects of spray liquid rebounding from the column wall and mist formation. The effective area would also be affected by the gas velocity which would tend to suspend the smaller particles in the column or remove them by entrainment at higher velocities. It was decided that any one of these problems in themselves were worthy of investigation without the added unknown effect of surface tension and surface active agents. The field of study was then shifted to liquid drops and the equipment was altered accordingly.

The observations of Hatta and Babba (22,23) and others (18,20) that the rate of absorption of gases by free falling liquid drops is greatest after the drop has just formed and begins to fall appeared to be logical. This may be affected by turbulence of the liquid surface due to the separation of the drop from the forming device. However, it appears more logical to this investigator that the observed result is a combination of two effects, the first of which is that the average driving force or concentration difference is greatest at this stage since one of the limits is the fact that the concentration of the gas in the liquid drop is essentially zero as the drop leaves the forming device. The second is that the average time of exposure is greater in this zone simply due to the fact that the drop is governed by the law of gravity and the velocity of fall is greater in the lower part of the column.

The results of this investigation indicate that a decrease in the surface tension of the liquid phase produced no change in the over-all liquid or gas film coefficient obtained for the absorption of the gases employed. An indirect effect was observed in that the number of liquid drops per minute increased with a decrease in surface tension. This produced an increase in the effective area for mass transfer as illustrated in Chapter VII, Methods of Calculation. The moles of gas absorbed per unit of time also increased with a decrease in the surface tension of the liquid. However, in the final analysis, these results offset each other in the calculation of the over-all mass transfer coefficient so that there was no change in this function.

Assuming the formation of a Gibbs surface under the conditions of this investigation, the resulting excess surface concentrations of Aersol OT can be calculated as illustrated in Section IV of the Appendix. It is seen that the amount of surface active agent removed from the interior of the drop to provide a Gibbs surface is negligible and that the contribution of the liquid bulk concentration to the surface coverage is negligible. Assuming that the effective cross-sectional area of Aersol OT is thirty square Angstrom units for the purpose of a sample calculation, the per cent of the surface covered by the Aersol OT molecules in the Gibbs surface can be computed. This is based upon the assumption that the Aersol OT molecules are completely oriented and are in a vertical position on the surface of the liquid. However, it should be noted that if a completely covered surface or compact film is not attained, then the actual area covered per molecule might be greater than the cross-sectional area of the molecule due to the resulting disordered arrangement of the

molecules upon the liquid surface. Thus we are unable to use the value of the Gibbs excess surface concentration in this case to calculate exactly the amount of surface covered by the surface active agent molecules because the amount of surface obscured per molecule may actually increase slightly as the surface concentration decreases due to the tendency of some of the molecules, so to speak, to topple over to a horizontal position. The amount of surface covered by the surface active agent would thus vary with each individual agent due to the structure of the molecule and the resulting structure of the Gibbs surface.

Consequently, if the Gibbs surface were assumed to be an important factor for consideration in gas absorption where the liquid film is controlling one would expect an initial decrease in the rate of absorption. This should then be attributed to an increase in the resistance of the liquid film due to the presence of the Gibbs surface and the resulting decrease in the effective area for mass transfer. This decrease in the rate of absorption should continue until the Gibbs excess surface concentration attains its maximum value. The absorption rate would then be expected to increase until the rate of absorption is approximately equal to the value obtained when no surface active agent is used as the value of the excess surface concentration tends to zero.

When the gas film is the controlling factor, some investigators (34,35,36) have found that use of surface active agents produce no effect upon the rate of gas absorption. However, if one again postulates that the Gibbs surface is a major consideration, there should be an observable change in the rate of absorption since the area available for mass transfer has been decreased and varies as previously outlined for the liquid

film. The effect upon the absorption rate would not be expected to be as great as in the case where the liquid film is controlling.

The literature survey indicates that a mono-molecular layer of an insoluble material on the surface of a liquid may cause an appreciable decrease in the rate of gas absorption. This film has different characteristics and properties from the Gibbs surface. Consequently, conclusions drawn from measurements with mono-molecular layers of insoluble materials should not be applied to systems where the surface active agents are soluble and the Gibbs surface is assumed to be attained. Therefore, arguments to the effect that since insoluble mono-molecular layers have been shown to decrease the rate of absorption, then the decrease in absorption rate found by some experimenters working with soluble surface active agents was due to the Gibbs surface, are not valid.

It has been pointed out that there exists no agreement among the previous investigators as to the effect of the addition of surface active agents upon gas absorption--some claiming an increase, some a decrease, and others no effect as the surface tension of the liquid phase is changed. The influence of lowered tension upon the absorption coefficient cannot be determined from these published results because the product of the absorption coefficient multiplied by the area was determined rather than the individual factors.

In this investigation the absorption coefficients were determined as well as the area, and it has been definitely shown that the addition of surface active agents does not effect the rate of absorption and, consequently, the Gibbs surface does not offer any significant resistance.

The divergent results of previous investigators have been explained

by taking into consideration such factors as, first, an increase in wetting and effective area for mass transfer with a decrease in surface tension in packed columns; and second, a decrease in the effective area for mass transfer and in turbulence with a decrease in surface tension in wetted wall columns.

CHAPTER IX

CONCLUSIONS

From the results of this investigation the following conclusions were drawn:

1. The over-all transfer coefficient for the absorption of a diffusing gas from a gas mixture by water droplets in a spray type column is not affected by the concentration of the diffusing gas in the main gas stream.

2. The over-all transfer coefficient for the absorption of a diffusing gas from a gas mixture by water droplets in a spray type column is not affected either by the surface tension of the absorbing liquid or the concentration of the surface active agents, such as Aersol OT, Sterox SK, and Victawet 12, used to alter the surface tension of the liquid.

3. The over-all transfer coefficient for the absorption of carbon dioxide and sulfur dioxide by water drops in a spray type column is not affected by the gas rate.

4. The over-all transfer coefficient for the absorption of ammonia by water droplets in a spray type column increases with an increase in gas velocity and may be correlated by an empirical equation as,

$$K_G = 0.213 Re^{0.25}$$

5. The Gibbs surface and the excess concentration of surface active agents in this surface have been shown to offer negligible resistance to mass transfer.

APPENDIX

APPENDIX I

Table I. Surface Tension Measurements

	Concentration wt. %	Surface Tension dynes/cm.	Surface conc. lb-moles/sq. ft.
A. Aerosol OT	0.001	64.3	
	0.002	58.9	
	0.003		3.9×10^{-10}
	0.004	49.5	
	0.005	46.4	
	0.01	42.2	
	0.012		5.9×10^{-10}
	0.015	40.3	
	0.02	38.8	
	0.04	34.0	6.2×10^{-10}
	0.06	30.9	6.03×10^{-10}
	0.1	28.2	3.12×10^{-10}
	0.15	26.9	
	0.2	26.7	
B. Victawet 12	0.0002	61.2	
	0.0003	54.4	2.3×10^{-10}
	0.0004	50.1	
	0.0005	46.8	
	0.001	41.7	
	0.002	38.6	3.42×10^{-10}
	0.004	35.2	3.78×10^{-10}
	0.006	33.3	
	0.008	31.8	4.15×10^{-10}
	0.01	30.4	4.22×10^{-10}
	0.02	28.6	

(Continued)

Table I. Surface Tension Measurements (concluded)

	Concentration wt. %	Surface Tension dynes/cm.	Surface conc. lb-moles/sq. ft.
C. Sterox SK	0.0001	69.7	
	0.0005	65.4	
	0.001	60.7	
	0.0025	51.7	8.46×10^{-10}
	0.003	49.2	
	0.005	43.3	9.8×10^{-10}
	0.01	34.3	10.6×10^{-10}
	0.015	31.6	6.1×10^{-10}
	0.02	29.5	4.66×10^{-10}
	0.03	27.5	
	0.05	25.6	
	0.1	25.2	

Table II. Absorption of Carbon Dioxide

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg.	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
1	0.00	72.4	742	79	100	1.69	30.04	5.57	7.22 - 7.19
2						3.38	60.08		7.22 - 7.26
3						5.07	90.12		7.19 - 7.22
4						6.76	120.16		7.19 - 7.22
5					80	6.89	117.40		7.22 - 7.22
6						13.79	234.80		7.19 - 7.26
7						20.68	352.20		7.26 - 7.22
8						27.57	469.60		7.22 - 7.26
9					65	3.01	49.37		7.23 - 7.17
10						6.02	98.75		7.20 - 7.20
11						9.02	148.12		7.22 - 7.17
12						12.03	197.49		7.17 - 7.23
13	S-0.2	25.15	745	80	100	1.69	30.04	4.11	7.28 - 7.27
14						3.38	60.08		7.26 - 7.25
15						5.07	90.12		7.28 - 7.26
16						6.76	120.16		7.27 - 7.27
17					80	6.89	117.40		7.29 - 7.33
18						13.79	234.80		7.29 - 7.39
19						20.68	352.20		7.28 - 7.32
20						27.57	469.60		7.30 - 7.31
21					65	3.01	49.37		7.38 - 7.41
22						6.02	98.75		7.42 - 7.39
23						9.03	148.12		7.38 - 7.42

(Continued)

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
24						12.03	197.49		7.39 - 7.39
25	S-0.1	25.20	745	80	100	1.69	30.04	4.11	7.25 - 7.26
26						3.38	60.08		7.27 - 7.27
27						5.07	90.12		7.29 - 7.26
28						6.76	120.16		7.28 - 7.27
29					80	6.89	117.40		7.29 - 7.33
30						13.79	234.80		7.33 - 7.36
31						20.68	352.20		7.29 - 7.31
32						27.57	469.60		7.32 - 7.29
33					65	3.01	49.37		7.42 - 7.37
34						6.02	98.75		7.39 - 7.36
35						9.03	148.12		7.43 - 7.39
36						12.03	197.49		7.39 - 7.39
37	S-.075	25.30	745	81	100	1.69	30.04	4.11	7.23 - 7.18
38						3.38	60.08		7.19 - 7.19
39						5.07	90.12		7.15 - 7.19
40						6.76	120.16		7.20 - 7.17
41					80	6.89	117.40		7.23 - 7.17
42						13.79	234.80		7.19 - 7.25
43						20.68	352.20		7.22 - 7.21
44						27.57	469.60		7.17 - 7.23
45					65	3.01	49.37		7.30 - 7.35
46						6.02	98.75		7.30 - 7.30

(Continued)

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F.	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
47						9.03	148.12		7.41 - 7.23
48						12.03	197.49		7.38 - 7.29
49	S-.05	25.60	742	79	100	1.69	30.04	4.11	7.32 - 7.35
50						3.38	60.08		7.37 - 7.37
51						5.07	90.12		7.35 - 7.38
52						6.76	120.16		7.38 - 7.35
53					80	6.89	117.40		7.40 - 7.35
54						13.79	234.80		7.35 - 7.39
55						20.68	352.20		7.38 - 7.38
56						27.57	469.60		7.40 - 7.35
57					65	3.01	49.37		7.50 - 7.45
58						6.02	98.75		7.53 - 7.49
59						9.03	148.12		7.47 - 7.51
60						12.03	197.49		7.49 - 7.49
61	S-.01	34.30	742	79	100	1.69	30.04	4.91	7.39 - 7.45
62						3.38	60.08		7.43 - 7.43
63						5.07	90.12		7.36 - 7.40
64						6.76	120.16		7.41 - 7.40
65					80	6.89	117.40		7.35 - 7.36
66						13.79	234.80		7.36 - 7.42
67						20.68	352.20		7.42 - 7.37
68						27.57	469.60		7.37 - 7.39
69					65	3.01	49.37		7.42 - 7.52
70						6.02	98.75		7.52 - 7.44
71						9.03	148.12		7.45 - 7.45

(Continued)

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
72						12.03	197.49		7.50 - 7.43
73	S-.003	49.20	742	80	100	1.69	30.04	5.43	7.38 - 7.43
74						2.38	60.08		7.39 - 7.44
75						5.07	90.12		7.40 - 7.40
76						6.76	120.16		7.40 - 7.44
77					80	6.89	117.40		7.22 - 7.27
78						13.79	234.80		7.27 - 7.31
79						20.68	352.20		7.31 - 7.25
80						27.57	469.60		7.24 - 7.29
81					65	3.01	49.37		7.33 - 7.26
82						6.02	98.75		7.28 - 7.28
83						9.03	148.12		7.30 - 7.25
84						12.03	197.49		7.31 - 7.25
85	S-.001	60.70	742	81	100	1.69	30.04	5.51	7.30 - 7.36
86						3.38	60.08		7.31 - 7.31
87						5.07	90.12		7.35 - 7.30
88						6.76	120.16		7.35 - 7.31
89					80	6.89	117.40		7.26 - 7.19
90						13.79	234.80		7.24 - 7.20
91						20.68	352.20		7.22 - 7.26
92						27.57	469.60		7.18 - 7.23
93					65	3.01	49.37		7.18 - 7.14
94						6.02	98.75		7.21 - 7.12
95						9.03	148.12		7.13 - 7.13
96						12.03	197.49		7.09 - 7.12

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
97	V-0.2	26.30	735	79	100	1.69	30.04	4.58	7.40 - 7.36
98						3.38	60.08		7.44 - 7.43
99						5.07	90.12		7.41 - 7.40
100						6.76	120.16		7.43 - 7.39
101					80	6.89	117.40		7.47 - 7.43
102						13.79	234.80		7.46 - 7.41
103						20.68	352.20		7.47 - 7.41
104						27.57	469.60		7.45 - 7.44
105					65	3.01	49.37		7.41 - 7.48
106						6.02	98.75		7.39 - 7.45
107						9.03	148.12		7.44 - 7.45
108						12.03	197.49		7.49 - 7.43
109	V-.075	27.90	735	79	100	1.69	30.04	4.88	7.38 - 7.43
110						3.38	60.08		7.40 - 7.34
111						5.07	90.12		7.40 - 7.40
112						6.76	120.16		7.43 - 7.37
113					80	6.89	117.40		7.39 - 7.46
114						13.79	234.80		7.43 - 7.44
115						20.68	352.20		7.41 - 7.48
116						27.57	469.60		7.47 - 7.42
117					65	3.01	49.37		7.43 - 7.37
118						6.02	98.75		7.48 - 7.41
119						9.03	148.12		7.40 - 7.41
120						12.03	197.49		7.45 - 7.38

(Continued)

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F.	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
121	V-.02	23.60	735	80	100	1.69	30.04	5.13	7.33 - 7.38
122						3.38	60.08		7.33 - 7.37
123						5.07	90.12		7.33 - 7.34
124						6.76	120.16		7.37 - 7.31
125					80	6.89	117.40		7.30 - 7.36
126						13.79	234.80		7.33 - 7.32
127						20.68	352.20		7.36 - 7.31
128						27.57	469.60		7.32 - 7.28
129					65	3.01	49.37		7.44 - 7.37
130						6.02	98.75		7.39 - 7.51
131						9.03	148.12		7.42 - 7.39
132						12.03	197.49		7.43 - 7.43
133	V-.01	30.40	742	79	100	1.69	30.04	5.24	7.26 - 7.30
134						3.38	60.08		7.34 - 7.29
135						5.07	90.12		7.30 - 7.33
136						6.76	120.16		7.28 - 7.33
137					80	6.89	117.40		7.37 - 7.43
138						13.79	234.80		7.39 - 7.40
139						20.68	352.20		7.36 - 7.42
140						27.57	469.60		7.34 - 7.41
141					65	3.01	49.37		7.19 - 7.25
142						6.02	98.75		7.16 - 7.25
143						9.03	148.12		7.21 - 7.20
144						12.03	197.49		7.21 - 7.14

(Continued)

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
145	V-.005	34.30	742	80	100	1.69	30.04	5.39	7.29 - 7.32
146						3.38	60.08		7.25 - 7.33
147						5.07	90.12		7.29 - 7.30
148						6.76	120.16		7.33 - 7.29
149					80	6.89	117.40		7.25 - 7.31
150						13.79	234.80		7.30 - 7.28
151						20.68	352.20		7.30 - 7.31
152						27.57	469.60		7.28 - 7.30
153					65	3.01	49.37		7.24 - 7.19
154						6.02	98.75		7.12 - 7.21
155						9.03	148.12		7.20 - 7.29
156						12.03	197.49		7.24 - 7.21
157	V-.001	41.70	742	80	100	1.69	30.04	5.43	7.31 - 7.28
158						3.38	60.08		7.26 - 7.33
159						5.07	90.12		7.36 - 7.29
160						6.76	120.16		7.30 - 7.31
161					80	6.89	117.40		7.35 - 7.29
162						13.79	234.80		7.27 - 7.25
163						20.68	352.20		7.31 - 7.22
164						27.57	469.60		7.28 - 7.29
165					65	3.01	49.37		7.14 - 7.17
166						6.02	98.75		7.15 - 7.24
167						9.03	148.12		7.24 - 7.18
168						12.03	197.49		7.23 - 7.19

(Continued)

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
169	V-.0004	50.10	742	81	100	1.69	30.04	5.46	7.37 - 7.31
170						3.38	60.08		7.42 - 7.35
171						5.07	90.12		7.34 - 7.33
172						6.76	120.16		7.29 - 7.32
173					80	6.89	117.40		7.30 - 7.39
174						13.79	234.80		7.35 - 7.33
175						20.68	352.20		7.27 - 7.31
176						27.57	469.60		7.32 - 7.30
177					65	3.01	49.37		7.36 - 7.31
178						6.02	98.75		7.27 - 7.23
179						9.03	148.12		7.23 - 7.29
180						12.03	197.49		7.25 - 7.25
181	A-.300	26.60	742	79	100	1.69	30.04	4.11	7.39 - 7.35
182						3.38	60.08		7.35 - 7.35
183						5.07	90.12		7.39 - 7.35
184						6.76	120.16		7.36 - 7.29
185					80	6.89	117.40		7.38 - 7.38
186						13.79	234.80		7.32 - 7.33
187						20.68	352.20		7.38 - 7.40
188						27.57	469.60		7.38 - 7.38
189					65	3.01	49.37		7.48 - 7.53
190						6.02	98.75		7.49 - 7.53
191						9.03	148.12		7.51 - 7.49
192						12.03	197.49		7.49 - 7.49

(Continued)

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press mm Hg	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
193	A-.200	26.70	742	79	100	1.69	30.04	4.11	7.31 - 7.34
194						3.38	60.08		7.35 - 7.38
195						5.07	90.12		7.35 - 7.38
196						6.76	120.16		7.36 - 7.36
197					80	6.89	117.40		7.42 - 7.37
198						13.79	234.80		7.37 - 7.37
199						20.68	352.20		7.40 - 7.40
200						27.57	469.60		7.33 - 7.31
201					65	3.01	49.37		7.53 - 7.46
202						6.02	98.75		7.46 - 7.50
203						9.03	148.12		7.47 - 7.47
204						12.03	197.49		7.52 - 7.47
205	A-.1	28.20	742	79	100	1.69	30.04	4.11	7.37 - 7.34
206						3.38	60.08		7.36 - 7.36
207						5.07	90.12		7.35 - 7.35
208						6.76	120.16		7.38 - 7.35
209					80	6.89	117.40		7.37 - 7.40
210						13.79	234.80		7.37 - 7.39
211						20.68	352.20		7.40 - 7.40
212						27.57	469.60		7.40 - 7.37
213					65	3.01	49.37		7.48 - 7.48
214						6.02	98.75		7.52 - 7.48
215						9.03	148.12		7.55 - 7.49
216						12.03	197.49		7.46 - 7.47

(Continued)

Table II. Absorption of Carbon Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
217	A-.075	29.90	738	81	100	1.69	30.04	4.16	7.50 - 7.51
218						3.38	60.08		7.52 - 7.49
219						5.07	90.12		7.50 - 7.48
220						6.76	120.16		7.51 - 7.48
221					80	6.89	117.40		7.62 - 7.60
222						13.79	234.80		7.58 - 7.61
223						20.68	352.20		7.63 - 7.60
224						27.57	469.60		7.61 - 7.61
225					65	3.01	49.37		7.31 - 7.36
226						6.02	98.75		7.36 - 7.36
227						9.03	148.12		7.34 - 7.38
228						12.03	197.49		7.34 - 7.35
229	A-.05	32.70	738	81	100	1.69	30.04	4.42	7.43 - 7.41
230						3.38	60.08		7.42 - 7.42
231						5.07	90.12		7.41 - 7.41
232						6.76	120.16		7.44 - 7.40
233					80	6.89	117.40		7.46 - 7.51
234						13.79	234.80		7.51 - 7.47
235						20.68	352.20		7.49 - 7.49
236						27.57	469.60		7.49 - 7.47
237					65	3.01	49.37		7.28 - 7.24
238						6.02	98.75		7.29 - 7.24
239						9.03	148.12		7.25 - 7.25
240						12.03	197.49		7.24 - 7.25

(Continued)

Table II. Absorption of Carbon Dioxide (concluded)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	CO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient
241	A-.01	42.20	742	80	100	1.69	30.04	5.11	7.42 - 7.38
242						3.38	60.08		7.40 - 7.40
243						5.07	90.12		7.39 - 7.42
244						6.76	120.16		7.41 - 7.37
245					80	6.89	117.40		7.33 - 7.33
246						13.79	234.80		7.36 - 7.32
247						20.68	352.20		7.36 - 7.35
248						27.57	469.60		7.32 - 7.36
249					65	3.01	49.37		7.32 - 7.28
250						6.02	98.75		7.29 - 7.30
251						9.03	148.12		7.29 - 7.34
252						12.03	197.49		7.30 - 7.29
253	A-.002	58.90	742	80	100	1.69	30.04	5.51	7.37 - 7.32
254						3.38	60.08		7.37 - 7.34
255						5.07	90.12		7.36 - 7.33
256						6.76	120.16		7.35 - 7.36
257					80	6.89	117.40		7.23 - 7.29
258						12.79	234.80		7.22 - 7.26
259						20.68	352.20		7.29 - 7.27
260						27.57	469.60		7.25 - 7.25
261					65	3.01	49.37		7.14 - 7.21
262						6.02	98.75		7.23 - 7.18
263						9.03	148.12		7.19 - 7.19
264						12.03	197.49		7.14 - 7.21

Table III. Absorption of Sulfur Dioxide

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
265	0.00	72.40	742	80	50	2.79	48.47	5.57	2.68 - 2.68
266						4.19	72.71		2.65 - 2.67
267						5.58	96.95		2.67 - 2.70
268						6.98	121.18		2.67 - 2.66
269					30	3.95	68.70		2.66 - 2.63
270						5.92	95.10		2.64 - 2.62
271						7.89	126.80		2.64 - 2.63
272						9.86	158.50		2.65 - 2.64
273					10	9.70	143.49		2.71 - 2.71
274						14.55	215.24		2.73 - 2.69
275						19.40	286.98		2.68 - 2.73
276						24.25	358.73		2.70 - 2.70
277	S-.2	25.15	742	82	50	2.79	48.47	4.11	2.75 - 2.73
278						4.19	72.71		2.72 - 2.74
279						5.58	96.95		2.74 - 2.75
280						6.98	121.18		2.74 - 2.72
281					30	3.95	68.40		2.70 - 2.70
282						5.92	95.10		2.70 - 2.68
283						7.89	126.80		2.68 - 2.69
284						9.86	158.50		2.70 - 2.69
285					10	9.70	143.49		2.70 - 2.71
286						14.55	215.24		2.73 - 2.81
287						19.40	286.98		2.81 - 2.82
288						24.25	358.73		2.72 - 2.73

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
289	S-.1	25.20	742	82	50	2.79	48.47	4.11	2.71 - 2.73
290						4.19	72.71		2.73 - 2.70
291						5.58	96.95		2.75 - 2.73
292						6.98	121.18		2.74 - 2.73
293					30	3.95	68.40		2.71 - 2.68
294						5.92	95.10		2.69 - 2.68
295						7.89	126.80		2.70 - 2.67
296						9.86	158.50		2.70 - 2.69
297					10	9.70	143.49		2.71 - 2.72
298						14.55	215.24		2.69 - 2.71
299						19.40	286.98		2.73 - 2.72
300						24.25	358.73		2.72 - 2.72
301	S-.075	25.30	742	82	50	2.79	48.47	4.11	2.74 - 2.73
302						4.19	72.71		2.73 - 2.74
303						5.58	96.95		2.74 - 2.72
304						6.98	121.18		2.74 - 2.73
305					30	3.95	68.40		2.69 - 2.69
306						5.92	95.10		2.69 - 2.67
307						7.89	126.80		2.71 - 2.69
308						9.86	158.50		2.70 - 2.68
309					10	9.70	143.49		2.73 - 2.74
310						14.55	215.24		2.72 - 2.72
311						19.40	286.98		2.71 - 2.72
312						24.25	358.73		2.70 - 2.81

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg.	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
313	S-.05	25.60	742	82	50	2.79	48.47	4.11	2.74 - 2.73
314						4.19	72.71		2.74 - 2.72
315						5.58	96.95		2.73 - 2.73
316						6.98	121.18		2.73 - 2.73
317					30	3.95	68.40		2.68 - 2.70
318						5.92	95.10		2.71 - 2.68
319						7.89	126.80		2.69 - 2.69
320						9.86	158.50		2.69 - 2.69
321					10	9.70	143.49		2.74 - 2.71
322						14.55	215.24		2.70 - 2.71
323						19.40	286.98		2.72 - 2.72
324						24.25	358.73		2.71 - 2.71
325	S-.01	34.30	735	82	50	2.79	48.47	4.91	2.65 - 2.64
326						4.19	72.71		2.65 - 2.67
327						5.58	96.95		2.65 - 2.66
328						6.98	121.18		2.66 - 2.64
329					30	3.95	68.40		2.67 - 2.69
330						5.92	95.10		2.67 - 2.66
331						7.89	126.80		2.65 - 2.68
332						9.86	158.50		2.67 - 2.67
333					10	9.70	143.49		2.71 - 2.71
334						14.55	215.24		2.72 - 2.70
335						19.40	286.98		2.74 - 2.72
336						24.25	358.73		2.73 - 2.73

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
337	S-.003	49.20	735	82	50	2.79	48.47	5.43	2.69 - 2.68
338						4.19	72.71		2.70 - 2.68
339						5.58	96.95		2.72 - 2.70
340						6.98	121.18		2.73 - 2.73
341					30	3.95	68.39		2.67 - 2.66
342						5.92	95.10		2.68 - 2.71
343						7.89	126.80		2.67 - 2.67
344						9.86	158.50		2.67 - 2.66
345					10	9.70	143.49		2.72 - 2.70
346						14.55	215.24		2.73 - 2.70
347						19.40	286.98		2.70 - 2.72
348						24.25	358.73		2.82 - 2.71
349	S-.001	60.70	735	82	50	2.79	48.47	5.51	2.70 - 2.68
350						4.19	72.71		2.66 - 2.62
351						5.58	96.95		2.62 - 2.66
352						6.98	121.18		2.62 - 2.61
353					30	3.95	68.40		2.66 - 2.64
354						5.92	95.10		2.66 - 2.65
355						7.89	126.80		2.67 - 2.64
356						9.86	158.50		2.64 - 2.64
357					10	9.70	143.49		2.70 - 2.70
358						14.55	215.24		2.68 - 2.65
359						19.40	286.98		2.70 - 2.68
360						24.25	358.73		2.68 - 2.68

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
361	A-.3	26.60	742	82	50	2.79	48.47	4.11	2.75 - 2.73
362						4.19	72.71		2.74 - 2.72
363						5.58	96.95		2.75 - 2.73
364						6.98	121.18		2.73 - 2.72
365					30	3.95	68.40		2.68 - 2.71
366						5.92	95.10		2.68 - 2.69
367						7.89	126.80		2.69 - 2.68
368						9.86	158.50		2.67 - 2.70
369					10	9.70	143.49		2.72 - 2.70
370						14.55	215.24		2.69 - 2.71
371						19.40	286.98		2.72 - 2.72
372						24.25	358.73		2.72 - 2.72
373	A-.2	26.70	742	82	50	2.79	48.47	4.11	2.75 - 2.73
374						4.19	72.71		2.72 - 2.74
375						5.58	96.95		2.72 - 2.74
376						6.98	121.18		2.75 - 2.73
377					30	3.95	68.40		2.71 - 2.69
378						5.92	95.10		2.68 - 2.71
379						7.89	126.80		2.70 - 2.69
380						9.86	158.50		2.69 - 2.69
381					10	9.70	143.49		2.71 - 2.71
382						14.55	215.24		2.74 - 2.72
383						19.40	286.98		2.69 - 2.71
384						24.25	358.73		2.73 - 2.74

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt.%	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
385	A-.1	28.20	742	82	50	2.79	48.47	4.11	2.74 - 2.72
386						4.19	72.71		2.75 - 2.71
387						5.58	96.95		2.74 - 2.73
388						6.98	121.18		2.74 - 2.73
389					30	3.95	68.40		2.67 - 2.69
390						5.92	95.10		2.69 - 2.70
391						7.89	126.80		2.69 - 2.69
392						9.86	158.50		2.70 - 2.68
393					10	9.70	143.49		2.73 - 2.74
394						14.55	215.24		2.72 - 2.72
395						19.40	286.98		2.70 - 2.72
396						24.25	358.73		2.71 - 2.71
397	A-.075	29.90	742	81	50	2.79	48.47	4.16	2.72 - 2.70
398						4.19	72.71		2.70 - 2.68
399						5.58	96.95		2.70 - 2.70
400						6.98	121.18		2.70 - 2.72
401					30	3.95	68.40		2.69 - 2.67
402						5.95	95.10		2.68 - 2.66
403						7.89	126.80		2.68 - 2.68
404						9.86	158.50		2.70 - 2.66
405					10	9.70	143.49		2.77 - 2.77
406						14.55	215.24		2.72 - 2.77
407						19.40	286.98		2.79 - 2.73
408						24.25	358.73		2.77 - 2.76

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
409	A-.05	32.70	742	81	50	2.79	48.47	4.42	2.70 - 2.72
410						4.19	72.71		2.73 - 2.68
411						5.58	96.95		2.70 - 2.70
412						6.98	121.18		2.71 - 2.68
413					30	3.95	68.40		2.67 - 2.68
414						5.92	95.10		2.67 - 2.67
415						7.89	126.80		2.66 - 2.68
416						9.86	158.50		2.66 - 2.67
417					10	9.70	143.49		2.75 - 2.75
418						14.55	215.24		2.78 - 2.74
419						19.40	286.98		2.73 - 2.75
420						24.25	358.73		2.75 - 2.73
421	A-.01	42.20	742	81	50	2.79	48.47	5.11	2.68 - 2.66
422						4.19	72.71		2.65 - 2.66
423						5.58	96.95		2.66 - 2.66
424						6.98	121.18		2.66 - 2.66
425					30	3.95	68.40		2.66 - 2.64
426						5.92	95.10		2.64 - 2.66
427						7.89	126.80		2.67 - 2.65
428						9.86	158.50		2.65 - 2.65
429					10	9.70	143.49		2.71 - 2.71
430						14.55	215.24		2.75 - 2.71
431						19.40	286.98		2.81 - 2.73
432						24.25	358.73		2.71 - 2.70

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
433	A-.002	58.90	742	81	50	2.79	48.47	5.51	2.69 - 2.68
434						4.19	72.71		2.71 - 2.69
435						5.58	96.95		2.69 - 2.69
436						6.98	121.18		2.71 - 2.68
437					30	3.95	68.40		2.62 - 2.59
438						5.92	95.10		2.58 - 2.61
439						7.89	126.80		2.61 - 2.64
440						9.86	158.50		2.61 - 2.61
441					10	9.70	143.49		2.71 - 2.71
442						14.55	215.24		2.69 - 2.68
443						19.40	286.98		2.65 - 2.72
444						24.25	358.73		2.69 - 2.69
445	VW-.2	26.30	740	82	50	2.79	48.47	4.58	2.73 - 2.74
446						4.19	72.71		2.76 - 2.72
447						5.58	96.95		2.73 - 2.71
448						6.98	121.18		2.73 - 2.73
449					30	3.95	68.40		2.68 - 2.67
450						5.92	95.10		2.69 - 2.68
451						7.89	126.80		2.31 - 2.32
452						9.86	158.50		2.33 - 2.31
453					10	9.70	143.49		2.80 - 2.74
454						14.55	215.24		2.76 - 2.76
455						19.40	286.98		2.74 - 2.78
456						24.25	358.73		2.75 - 2.76

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
457	VW-.075	27.90	740	82	50	2.79	48.47	4.88	2.68 - 2.70
458						4.19	72.71		2.67 - 2.68
459						5.58	96.95		2.67 - 2.68
460						6.98	121.18		2.68 - 2.68
461					30	3.95	68.40		2.71 - 2.70
462						5.92	95.10		2.70 - 2.70
463						7.89	126.80		2.68 - 2.71
464						9.86	158.50		2.71 - 2.70
465					10	9.70	143.49		2.77 - 2.76
466						14.55	215.24		2.80 - 2.74
467						19.40	286.98		2.76 - 2.76
468						24.25	358.73		2.79 - 2.74
469	VW-.02	28.60	740	82	50	2.79	48.47	5.13	2.69 - 2.68
470						4.19	72.71		2.69 - 2.67
471						5.58	96.95		2.68 - 2.68
472						6.98	121.18		2.68 - 2.68
473					30	3.95	68.40		2.71 - 2.70
474						5.92	95.10		2.71 - 2.71
475						7.89	126.80		2.72 - 2.74
476						9.86	158.50		2.71 - 2.70
477					10	9.70	143.49		2.77 - 2.75
478						14.55	215.24		2.78 - 2.75
479						19.40	286.98		2.76 - 2.76
480						24.25	358.73		2.78 - 2.75

(Continued)

Table III. Absorption of Sulfur Dioxide (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
481	VW-.01	30.40	740	82	50	2.79	48.47	5.24	2.71 - 2.71
482						4.19	72.71		2.73 - 2.70
483						5.58	96.95		2.71 - 2.70
484						6.98	121.18		2.71 - 2.70
485					30	3.95	68.40		2.73 - 2.72
486						5.92	95.10		2.73 - 2.73
487						7.89	126.80		2.74 - 2.71
488						9.86	158.50		2.73 - 2.71
489					10	9.70	143.49		2.81 - 2.78
490						14.55	215.24		2.79 - 2.80
491						19.40	286.98		2.79 - 2.79
492						24.25	358.73		2.83 - 2.79
493	VW-.005	34.30	740	82	50	2.79	48.47	5.39	2.69 - 2.70
494						4.19	72.71		2.68 - 2.70
495						5.58	96.95		2.70 - 2.70
496						6.98	121.18		2.68 - 2.69
497					30	3.95	68.40		2.71 - 2.73
498						5.92	95.10		2.70 - 2.72
499						7.89	126.80		2.71 - 2.72
500						9.86	158.50		2.73 - 2.76
501					10	9.70	143.49		2.76 - 2.78
502						14.55	215.24		2.74 - 2.78
503						19.40	286.98		2.76 - 2.79
504						24.25	358.73		2.78 - 2.76

(Continued)

Table III. Absorption of Sulfur Dioxide (concluded)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	SO_2 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_L
505	VW-.001	41.70	740	82	50	2.79	48.47	5.43	2.67 - 2.66
506						4.19	72.71		2.68 - 2.66
507						5.58	96.95		2.67 - 2.67
508						6.98	121.18		2.67 - 2.68
509					30	3.95	68.40		2.72 - 2.68
510						5.92	95.10		2.70 - 2.69
511						7.89	126.80		2.70 - 2.70
512						9.86	158.50		2.68 - 2.72
513					10	9.70	143.49		2.74 - 2.71
514						14.55	215.24		2.76 - 2.77
515						19.40	286.98		2.75 - 2.76
516						24.25	358.73		2.79 - 2.73
517	VW-.0004	50.10	740	82	50	2.79	48.47	5.46	2.68 - 2.70
518						4.19	72.71		2.69 - 2.70
519						5.58	96.95		2.69 - 2.69
520						6.98	121.18		2.69 - 2.68
521					30	3.95	68.40		2.71 - 2.72
522						5.92	95.10		2.74 - 2.71
523						7.89	126.80		2.70 - 2.74
524						9.86	158.50		2.72 - 2.72
525					10	9.70	143.49		2.74 - 2.77
526						14.55	215.24		2.78 - 2.81
527						19.40	286.98		2.76 - 2.79
528						24.25	358.73		2.73 - 2.78

Table IV. Absorption of Ammonia

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
529	0.00	72.40	742	82	5	96.41	1160.02	5.57	1.34 - 1.33
530						128.54	1546.66		1.41 - 1.42
531						160.68	1933.29		1.45 - 1.46
532						192.82	2320.05		1.56 - 1.57
533					3	54.07	775.90		1.10 - 1.07
534						108.15	1551.80		1.28 - 1.30
535						162.22	2327.69		1.48 - 1.51
536						216.29	3103.59		1.59 - 1.62
537					1	81.82	1396.91		1.46 - 1.47
538						163.64	2793.81		1.46 - 1.47
539						245.47	4190.72		1.61 - 1.62
540						327.29	5587.62		1.76 - 1.78
541	S-.2	25.15	742	82	5	96.41	1160.02	4.11	1.33 - 1.35
542						128.54	1546.66		1.42 - 1.41
543						160.68	1933.29		1.45 - 1.46
544						190.82	2320.05		1.55 - 1.57
545					3	54.07	775.90		1.08 - 1.09
546						108.15	1551.80		1.29 - 1.30
547						162.22	2327.60		1.50 - 1.53
548						216.29	3103.59		1.60 - 1.58
549						81.82	1396.91		1.23 - 1.24
550						163.64	2793.81		1.47 - 1.46
551						245.47	4190.72		1.61 - 1.62
552						327.29	5587.62		1.76 - 1.79

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
553	S-.1	25.20	742	82	5	96.41	1160.02	4.11	1.33 - 1.35
554						128.54	1546.66		1.41 - 1.41
555						160.68	1933.29		1.46 - 1.48
556						192.82	2320.05		1.56 - 1.59
557					3	54.07	775.90		1.08 - 1.09
558						108.15	1551.80		1.29 - 1.30
559						162.22	2327.60		1.53 - 1.50
560						216.29	3103.59		1.59 - 1.61
561					1	81.82	1396.91		1.23 - 1.24
562						163.64	2793.81		1.46 - 1.46
563						245.47	4190.72		1.61 - 1.62
564						327.29	5587.62		1.77 - 1.78
565	S-.075	25.30	742	82	5	96.41	1160.02	4.11	1.33 - 1.34
566						128.54	1546.66		1.42 - 1.41
567						160.68	1933.29		1.45 - 1.46
568						192.82	2320.05		1.45 - 1.47
569					3	54.07	775.90		1.08 - 1.09
570						108.15	1551.80		1.29 - 1.29
571						162.22	2327.69		1.49 - 1.51
572						216.29	3103.59		1.59 - 1.62
573					1	81.82	1396.91		1.23 - 1.25
574						163.64	2793.81		1.47 - 1.46
575						245.47	4190.72		1.60 - 1.62
576						327.29	5587.62		1.77 - 1.78

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
577	S-.050	25.60	742	82	5	96.41	1160.02	4.11	1.34 - 1.33
578						128.54	1546.66		1.42 - 1.43
579						160.68	1933.29		1.45 - 1.46
580						192.82	2320.05		1.57 - 1.55
581					3	54.07	775.90		1.10 - 1.08
582						108.15	1551.80		1.29 - 1.29
583						162.22	2327.69		1.53 - 1.49
584						216.29	3103.59		1.60 - 1.60
585					1	81.82	1396.91		1.24 - 1.24
586						163.64	2793.81		1.46 - 1.48
587						245.47	4190.72		1.60 - 1.62
588						327.29	5587.62		1.77 - 1.79
589	S-.01	34.30	742	82	5	96.41	1160.02	4.91	1.33 - 1.34
590						128.54	1546.66		1.42 - 1.41
591						160.68	1933.29		1.45 - 1.46
592						192.82	2320.05		1.56 - 1.58
593					3	54.07	775.90		1.08 - 1.10
594						108.15	1551.80		1.28 - 1.26
595						162.22	2327.69		1.48 - 1.52
596						216.29	3103.59		1.59 - 1.62
597					1	81.82	1396.91		1.24 - 1.25
598						163.64	2793.81		1.50 - 1.48
599						245.47	4190.72		1.63 - 1.61
600						327.29	5587.62		1.76 - 1.78

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
601	S-.003	49.20	742	82	5	96.41	1160.02	5.43	1.35 - 1.32
602						128.54	1546.66		1.40 - 1.39
603						160.68	1933.29		1.44 - 1.46
604						192.82	2320.05		1.54 - 1.58
605					3	54.07	775.90		1.06 - 1.09
606						108.15	1551.80		1.28 - 1.30
607						162.22	2327.69		1.49 - 1.51
608						216.29	3103.59		1.57 - 1.62
609					1	81.82	1396.91		1.22 - 1.25
610						163.64	2793.81		1.48 - 1.45
611						245.47	4190.72		1.67 - 1.63
612						327.29	5587.62		1.76 - 1.78
613	S-.001	60.70	742	82	5	96.41	1160.02	5.51	1.32 - 1.36
614						128.54	1546.66		1.40 - 1.43
615						160.68	1933.29		1.43 - 1.47
616						192.82	2320.05		1.55 - 1.59
617					3	54.07	775.90		1.08 - 1.09
618						108.15	1551.80		1.35 - 1.28
619						162.22	2327.69		1.50 - 1.48
620						216.29	3103.59		1.58 - 1.62
621					1	81.82	1396.91		1.22 - 1.25
622						163.64	2793.81		1.45 - 1.48
623						245.47	4190.72		1.60 - 1.63
624						327.29	5587.62		1.75 - 1.80

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
625	VW-.2	26.30	742	84	5	96.41	1160.02	4.58	1.35 - 1.32
626						128.54	1546.66		1.43 - 1.41
627						160.68	1933.29		1.47 - 1.44
628						192.82	2320.05		1.56 - 1.59
629					3	54.07	775.90		1.10 - 1.09
630						108.15	1551.80		1.30 - 1.29
631						162.22	2327.69		1.57 - 1.49
632						216.29	3103.59		1.59 - 1.63
633					1	81.82	1396.91		1.25 - 1.24
634						163.64	2793.81		1.47 - 1.48
635						245.47	4190.72		1.61 - 1.64
636						327.29	5587.62		2.58 - 2.21
637	VW-.075	27.90	742	84	5	96.41	1160.02	4.88	1.33 - 1.36
638						128.54	1546.66		1.40 - 1.43
639						160.68	1933.29		1.45 - 1.47
640						192.82	2320.05		1.55 - 1.59
641					3	54.07	775.90		1.09 - 1.10
642						108.15	1551.80		1.29 - 1.31
643						162.22	2327.69		1.49 - 1.53
644						216.29	3103.59		1.60 - 1.63
645					1	81.82	1396.91		1.25 - 1.25
646						163.64	2793.81		1.49 - 1.47
647						245.47	4190.72		1.64 - 1.62
648						327.29	5587.62		1.81 - 1.84

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
649	VW-.02	28.60	742	84	5	96.41	1160.02	5.13	1.34 - 1.35
650						128.54	1546.66		1.44 - 1.40
651						160.68	1933.29		1.47 - 1.45
652						192.82	2320.05		1.56 - 1.59
653					3	54.07	775.90		1.10 - 1.09
654						108.15	1551.80		1.30 - 1.30
655						162.22	2327.69		1.52 - 1.49
656						216.29	3103.59		1.59 - 1.63
657					1	81.82	1396.91		1.24 - 1.26
658						163.64	2793.81		1.45 - 1.47
659						245.47	4190.72		1.63 - 1.64
660						327.29	5587.62		1.73 - 1.77
661	VW-.01	30.40	742	84	5	96.41	1160.02	5.24	1.33 - 1.36
662						128.54	1546.66		1.41 - 1.43
663						160.68	1933.29		1.44 - 1.47
664						192.82	2320.05		1.56 - 1.58
665					3	54.07	775.90		1.08 - 1.10
666						108.15	1551.80		1.30 - 1.29
667						162.22	2327.69		1.49 - 1.52
668						216.29	3103.59		1.59 - 1.63
669					1	81.82	1396.91		1.24 - 1.26
670						163.64	2793.81		1.48 - 1.46
671						245.47	4190.72		1.62 - 1.62
672						327.29	5587.62		1.77 - 1.80

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
673	VW-.001	34.30	740	83	5	96.41	1160.02	5.43	1.32 - 1.35
674						128.54	1546.66		1.41 - 1.41
675						160.68	1933.29		1.43 - 1.47
676						192.82	2320.05		1.55 - 1.58
677					3	54.07	775.90		1.10 - 1.08
678						108.15	1551.80		1.29 - 1.29
679						162.22	2327.69		1.48 - 1.51
680						216.29	3103.59		1.59 - 1.63
681					1	81.82	1396.91		1.23 - 1.25
682						163.64	2793.81		1.46 - 1.49
683						245.47	4190.72		1.63 - 1.62
684						327.29	5587.62		1.76 - 1.80
685	VW-.005	41.70	740	83	5	96.41	1160.02	5.39	1.32 - 1.35
686						128.54	1546.66		1.43 - 1.39
687						160.68	1933.29		1.47 - 1.44
688						192.82	2320.05		1.55 - 1.58
689					3	54.07	775.90		1.09 - 1.09
690						108.15	1551.80		1.30 - 1.28
691						162.22	2327.69		1.48 - 1.52
692						216.29	3103.59		1.59 - 1.63
693					1	81.82	1396.91		1.25 - 1.24
694						163.64	2793.81		1.47 - 1.46
695						245.47	4190.72		1.62 - 1.62
696						327.29	5587.62		1.77 - 1.80

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
697	VW-.0004	50.10	740	83	5	96.41	1160.02	5.46	1.34 - 1.34
698						128.54	1546.66		1.40 - 1.43
699						160.68	1933.29		1.44 - 1.47
700						192.82	2320.05		1.55 - 1.58
701					3	54.07	775.90		1.08 - 1.09
702						108.15	1551.80		1.30 - 1.28
703						162.22	2327.69		1.49 - 1.51
704						216.29	3103.59		1.59 - 1.63
705					1	81.82	1396.91		1.24 - 1.25
706						163.64	2793.81		1.48 - 1.45
707						245.47	4190.72		1.64 - 1.60
708						327.29	5587.62		1.79 - 1.78
709	A-.3	26.60	742	82	5	96.41	1160.02	4.11	1.33 - 1.36
710						128.54	1546.66		1.42 - 1.42
711						160.68	1933.29		1.46 - 1.46
712						192.82	2320.05		1.55 - 1.57
713					3	54.07	775.90		1.08 - 1.09
714						108.15	1551.80		1.29 - 1.30
715						162.22	2327.69		1.50 - 1.52
716						216.29	3103.59		1.59 - 1.62
717					1	81.82	1396.91		1.24 - 1.23
718						163.64	2793.81		1.47 - 1.46
719						245.47	4190.72		1.61 - 1.63
720						327.29	5587.62		1.79 - 1.78

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
721	A-.2	26.70	742	82	5	96.41	1160.02	4.11	1.34 - 1.33
722						128.54	1546.66		1.40 - 1.43
723						160.68	1933.29		1.45 - 1.46
724						192.82	2320.05		1.55 - 1.57
725					3	54.07	775.90		1.09 - 1.09
726						108.15	1551.80		1.28 - 1.29
727						162.22	2327.69		1.49 - 1.50
728						216.29	3103.59		1.58 - 1.61
729						81.82	1396.91		1.24 - 1.25
730						163.64	2793.81		1.46 - 1.47
731						245.47	4190.72		1.62 - 1.61
732						327.29	5587.62		1.76 - 1.78
733	A-.1	28.20	742	82	5	96.41	1160.02	4.11	1.34 - 1.34
734						128.54	1546.66		1.49 - 1.41
735						160.68	1933.29		1.46 - 1.48
736						192.82	2320.05		1.57 - 1.56
737					3	54.07	775.90		1.09 - 1.08
738						108.15	1551.80		1.30 - 1.29
739						162.22	2327.69		1.51 - 1.55
740						216.29	3103.59		1.61 - 1.59
741					1	81.82	1396.91		1.23 - 1.24
742						163.64	2793.81		1.47 - 1.46
743						245.47	4190.72		1.58 - 1.62
744						327.29	5587.62		1.77 - 1.78

(Continued)

Table IV. Absorption of Ammonia (continued)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
745	A-.075	29.90	740	83	5	96.41	1160.02	4.16	1.32 - 1.35
746						128.54	1546.66		1.43 - 1.40
747						160.68	1933.29		1.44 - 1.47
748						192.82	2320.05		1.55 - 1.59
749					3	54.07	775.90		1.10 - 1.08
750						108.15	1551.80		1.28 - 1.31
751						162.22	2327.69		1.49 - 1.52
752						216.29	3103.59		1.58 - 1.62
753					1	81.82	1396.91		1.24 - 1.26
754						163.64	2793.81		1.46 - 1.50
755						245.47	4190.72		1.64 - 1.60
756						327.29	5587.62		1.76 - 1.81
757	A-.05	32.70	740	83	5	96.41	1160.02	4.42	1.32 - 1.35
758						128.54	1546.66		1.40 - 1.43
759						160.68	1933.29		1.44 - 1.47
760						192.82	2320.05		1.55 - 1.58
761					3	54.07	775.90		1.08 - 1.10
762						108.15	1551.80		1.28 - 1.30
763						162.22	2327.69		1.49 - 1.52
764						216.29	3103.59		1.58 - 1.62
765					1	81.82	1396.91		1.23 - 1.26
766						163.64	2793.81		1.46 - 1.49
767						245.47	4190.72		1.61 - 1.64
768						327.29	5587.62		1.76 - 1.80

(Continued)

Table IV. Absorption of Ammonia (concluded)

Run No.	Agent Wt. %	Surface Tension dynes/cm.	Press. mm Hg	Temp. °F	NH_3 %	Gas Rate G	Gas Rate $(Re)_G$	Drop Size mm	Absorption Coefficient K_G
769	A-.01	42.20	740	83	5	96.41	1160.02	5.11	1.32 - 1.35
770						128.54	1546.66		1.40 - 1.43
771						160.68	1933.29		1.45 - 1.46
772						192.82	2320.05		1.55 - 1.58
773					3	54.07	775.90		1.09 - 1.08
774						108.15	1551.80		1.29 - 1.30
775						162.22	2327.69		1.49 - 1.52
776						216.29	3103.59		1.59 - 1.63
777					1	81.82	1396.91		1.24 - 1.26
778						163.64	2793.81		1.47 - 1.47
779						245.47	4190.72		1.61 - 1.65
780						327.29	5587.62		1.77 - 1.81
781	A-.002	58.90	740	83	5	96.41	1160.02	5.51	1.34 - 1.32
782						128.54	1546.66		1.40 - 1.43
783						160.68	1933.29		1.46 - 1.44
784						192.82	2320.05		1.54 - 1.58
785					3	54.07	775.90		1.10 - 1.12
786						108.15	1556.80		1.28 - 1.30
787						162.22	2327.69		1.51 - 1.48
788						216.29	3103.59		1.58 - 1.61
789					1	81.82	1396.91		1.23 - 1.25
790						163.64	2793.81		1.47 - 1.47
791						245.47	4190.72		1.60 - 1.55
792						327.29	5587.62		1.76 - 1.80

NOMENCLATURE FOR TABLES II - IV

Run No.	Run number
% Wt. conc.	Per cent weight concentration of additive agent
A - Symbolizes Aersol OT S - Symbolizes Sterox SK V - Symbolizes Victawet 12 0.00 - Symbolizes no agent or plain tap water	
Surface tension	Surface tension of absorbing liquid in dynes/cm
Pressure	Prevailing atmospheric pressure in mm of mercury
Temperature	Temperature of inlet gas in degrees Fahrenheit
G	Gas mass flow rate in pounds per hour - square foot
Re_G	Reynold's number of the gas
K_L	Over-all absorption coefficient expressed in pound moles per hour-square foot- c, where c is the concentration in pound moles per cubic foot at 80 degrees Fahrenheit
K_G	Over-all absorption coefficient expressed in pound moles per hour-square foot-atmosphere at 82 degrees Fahrenheit

Note: The liquid rate was a constant for all runs at 0.5 pounds per minute.

The length of the drop fall was a constant throughout the investigation of 43.5 inches.

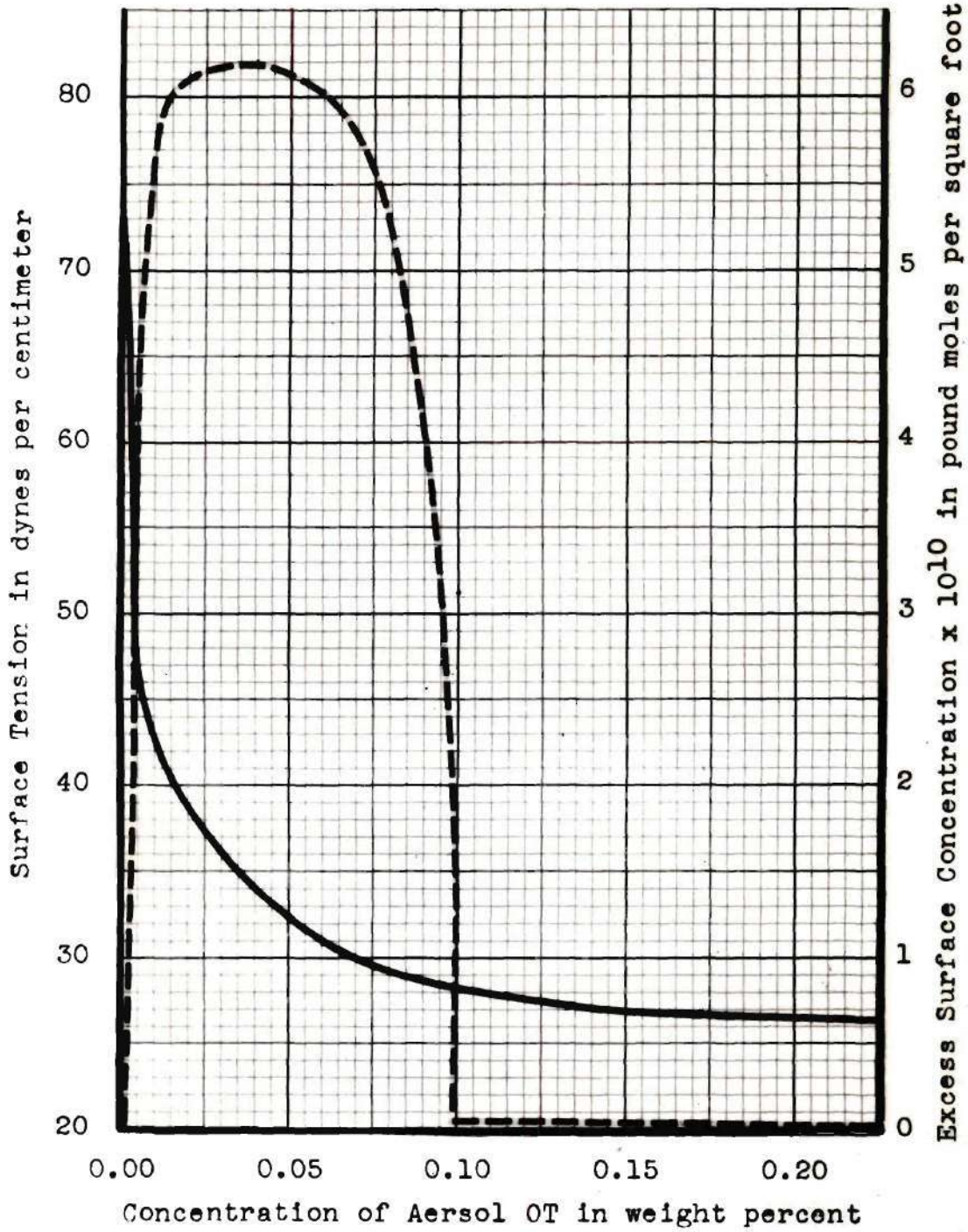


Figure 10. Surface Tension-Concentration Curve for Aersol OT

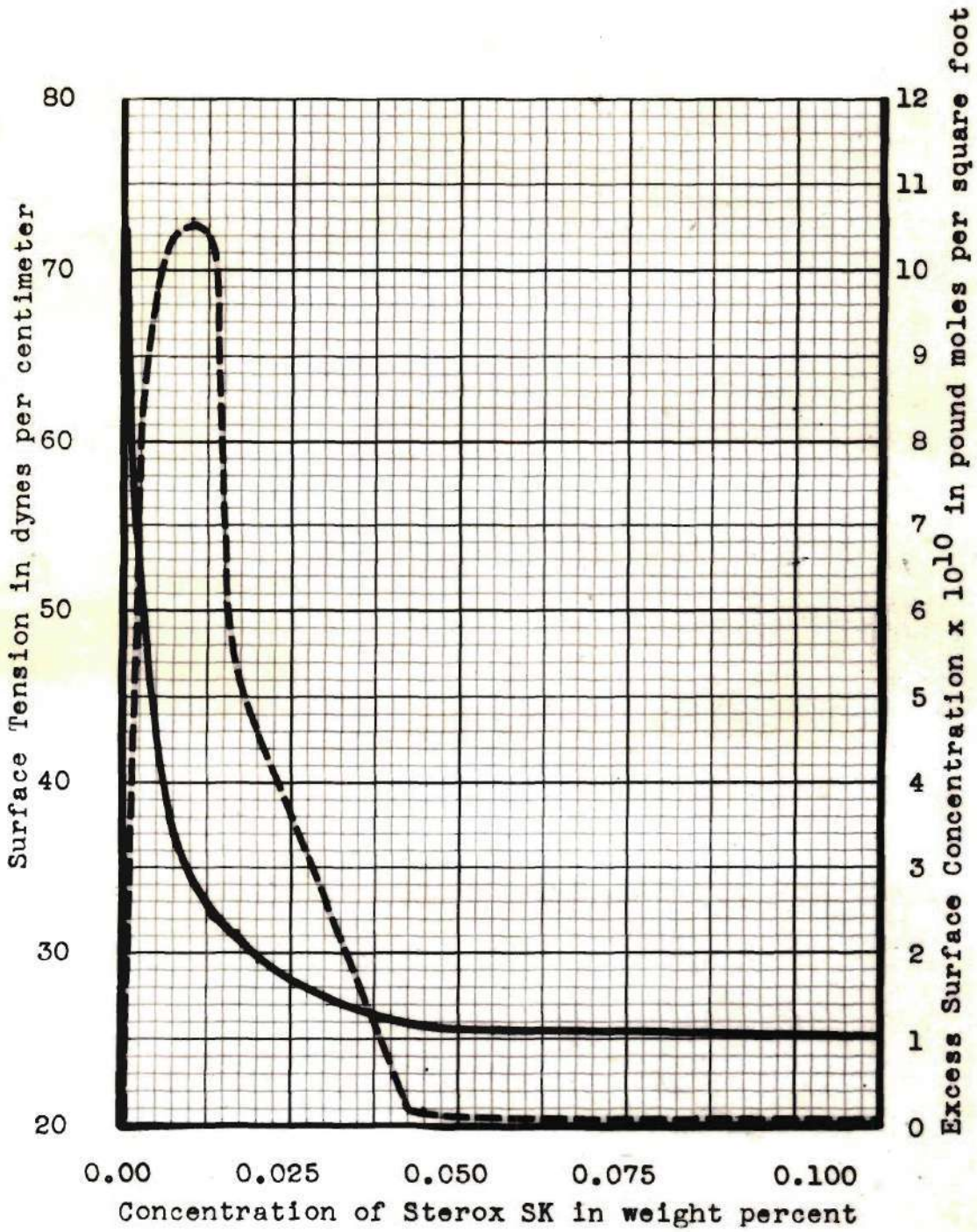


Figure 11. Surface Tension-Concentration Curve for Sterox SK

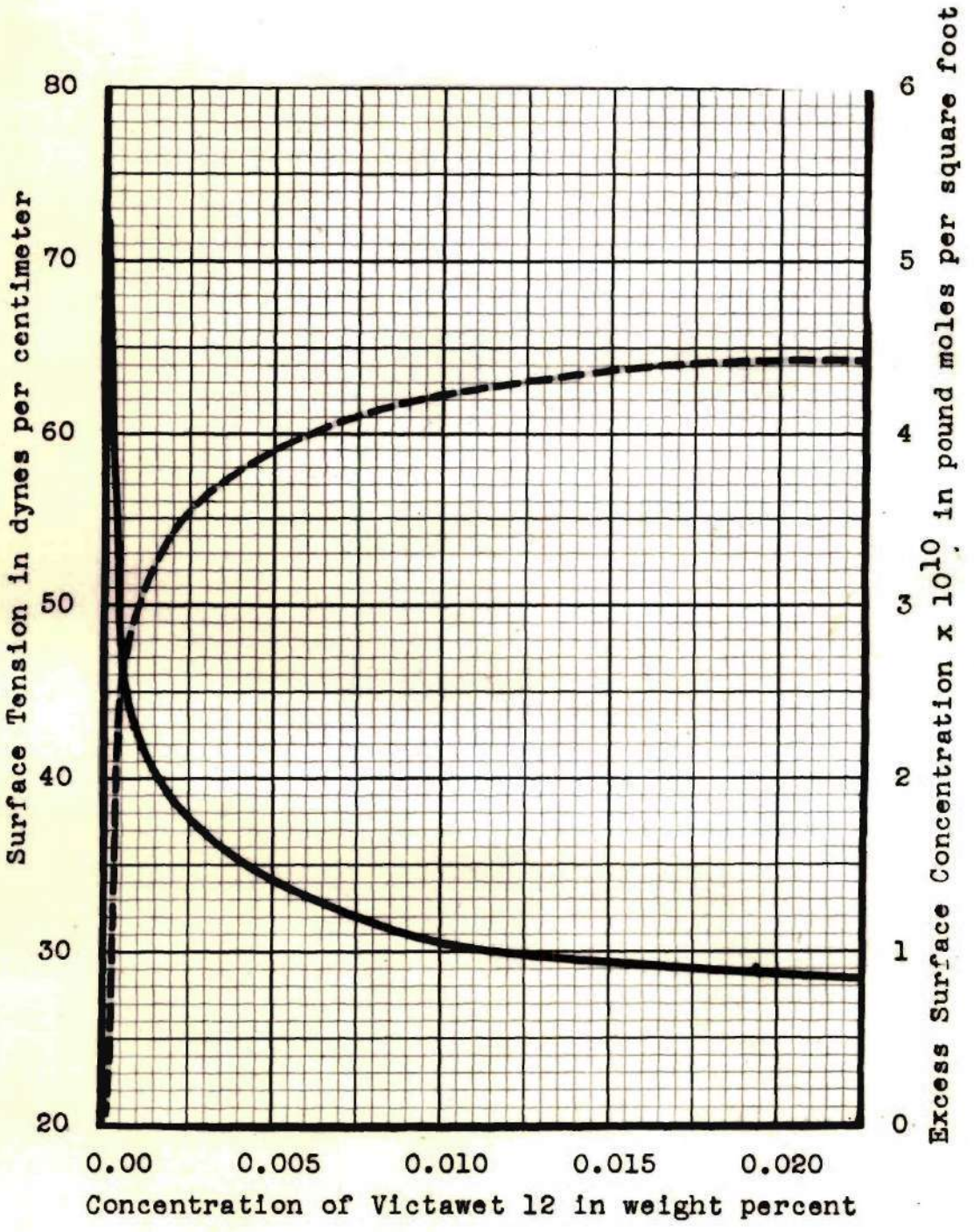


Figure 12. Surface Tension-Concentration Curve for Victawet 12

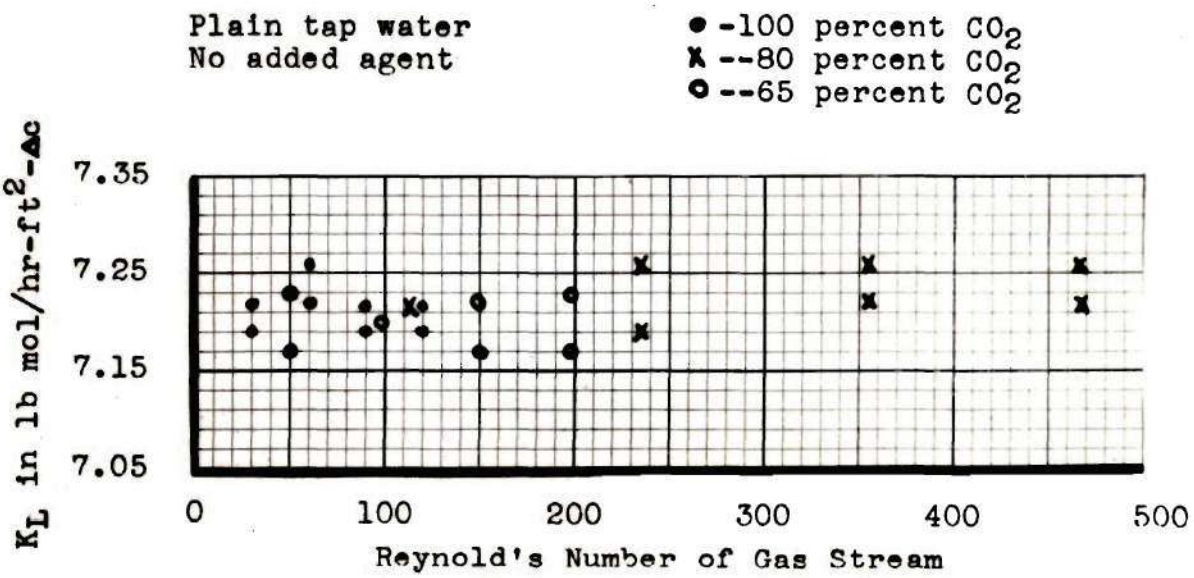


Figure 13. Absorption Coefficient for Carbon Dioxide versus Reynold's Number of Gas Stream

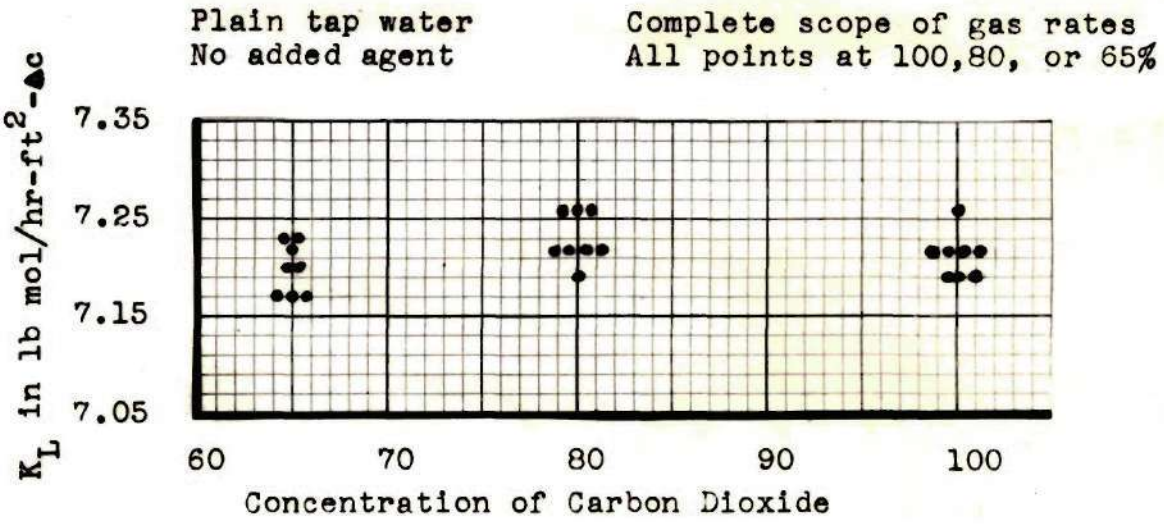


Figure 14. Absorption Coefficient for Carbon Dioxide versus Percent Concentration by Volume of Carbon Dioxide in Main Gas Stream

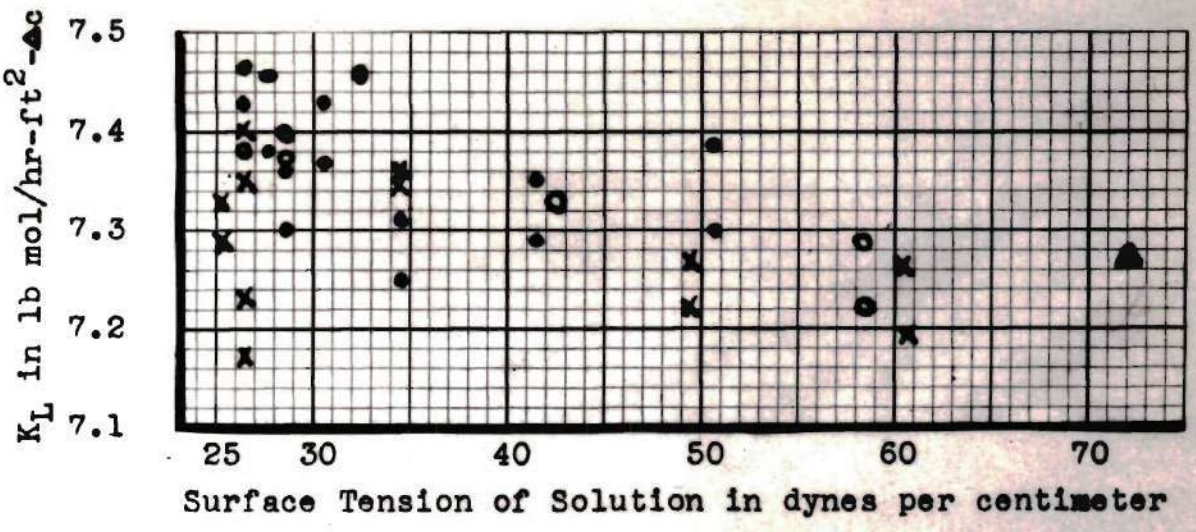


Figure 15. Absorption Coefficient for Carbon Dioxide versus Surface Tension of Absorbing Water

Both Figures illustrate data for concentration of 80% CO_2 Reynold's Number of gas, 117.40

○-Aersol OT
✗-Sterox SK
●-Victawet 12
▲-Tap water

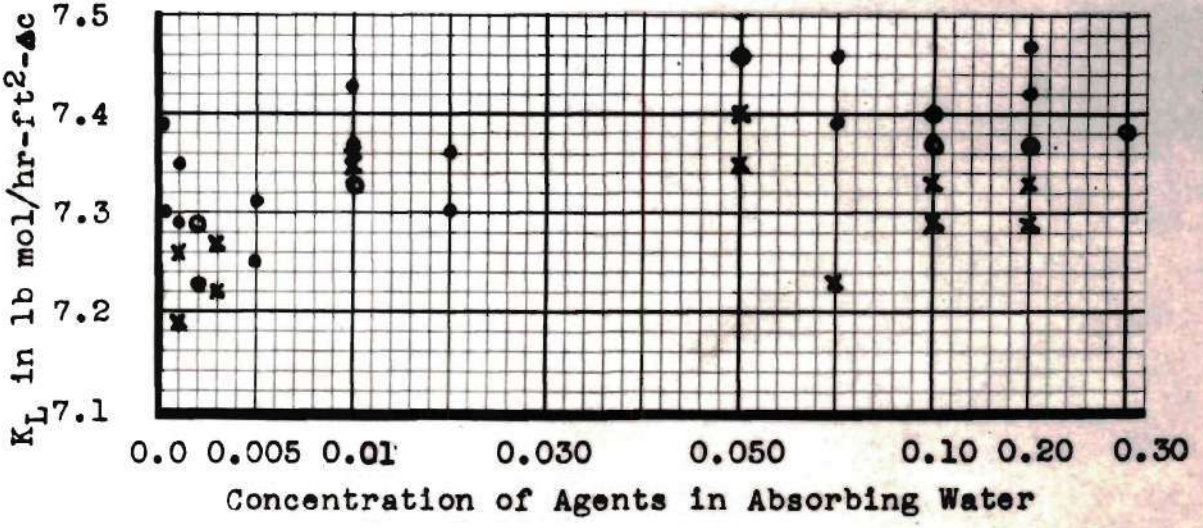


Figure 16. Absorption Coefficient for Carbon Dioxide versus Concentration of Agents in Absorbing Water

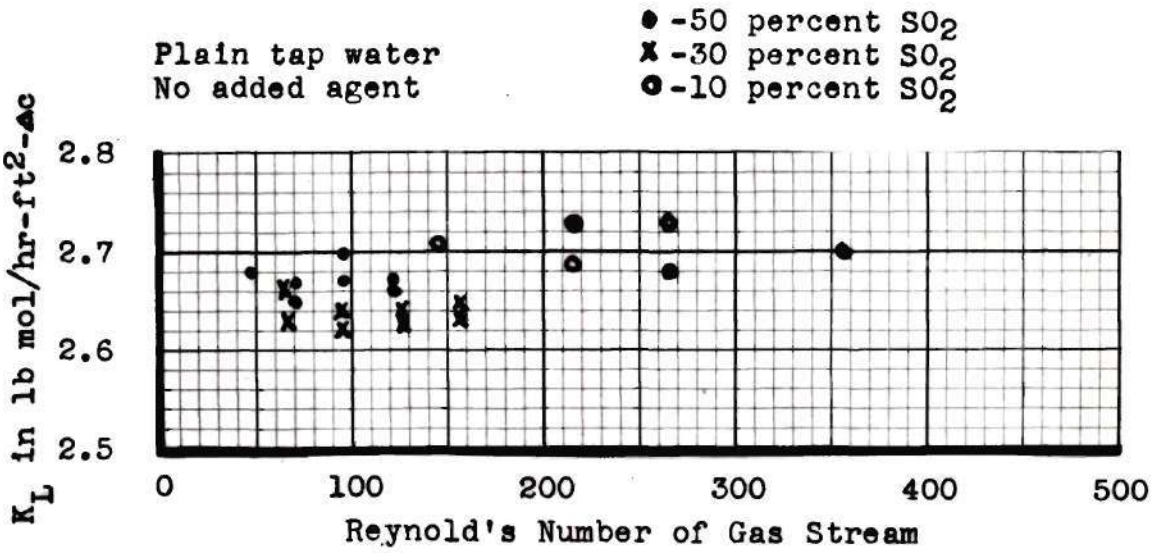


Figure 17. Absorption Coefficient for Sulfur Dioxide versus Reynold's Number of Gas Stream

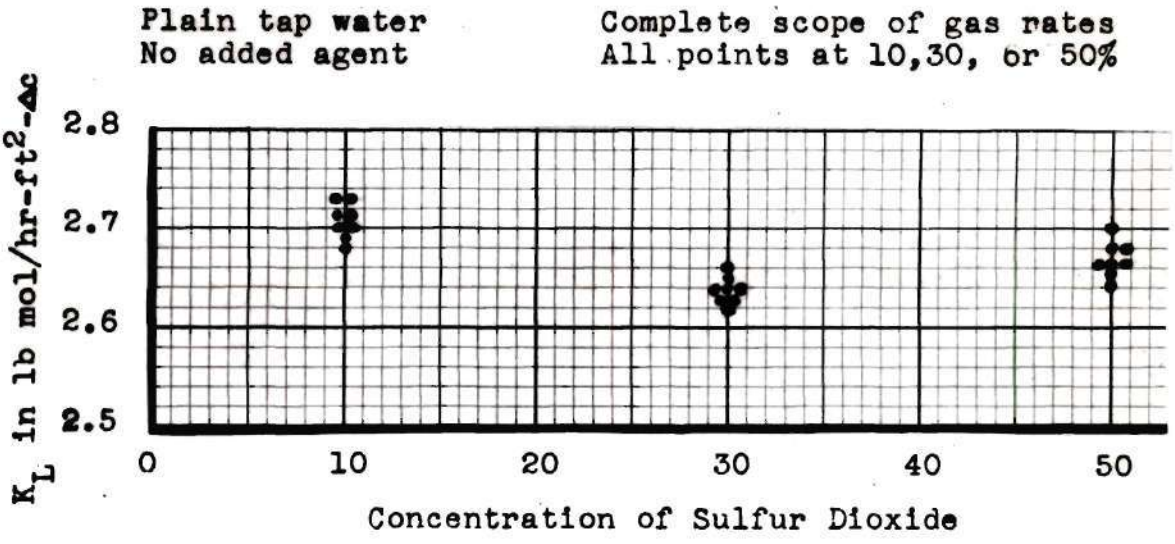


Figure 18. Absorption Coefficient for Sulfur Dioxide versus Percent Concentration by Volume of Sulfur Dioxide in Main Gas Stream

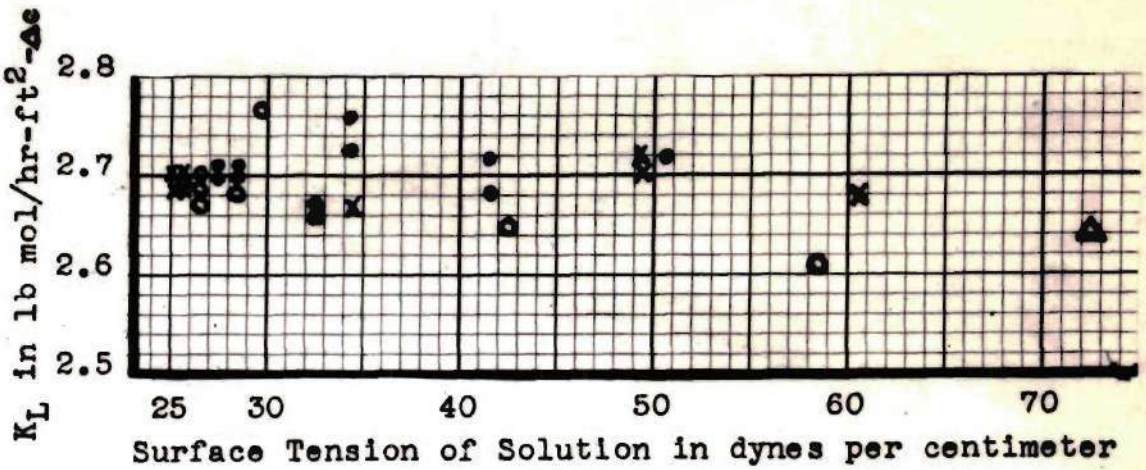


Figure 19. Absorption Coefficient for Sulfur Dioxide versus Surface Tension of Absorbing Water

Both Figures illustrate data for concentration of 30% SO_2 Reynold's Number of gas, 158.50

●-Aersol OT
✕-Sterox SK
●-Victawet 12
▲-Tap water

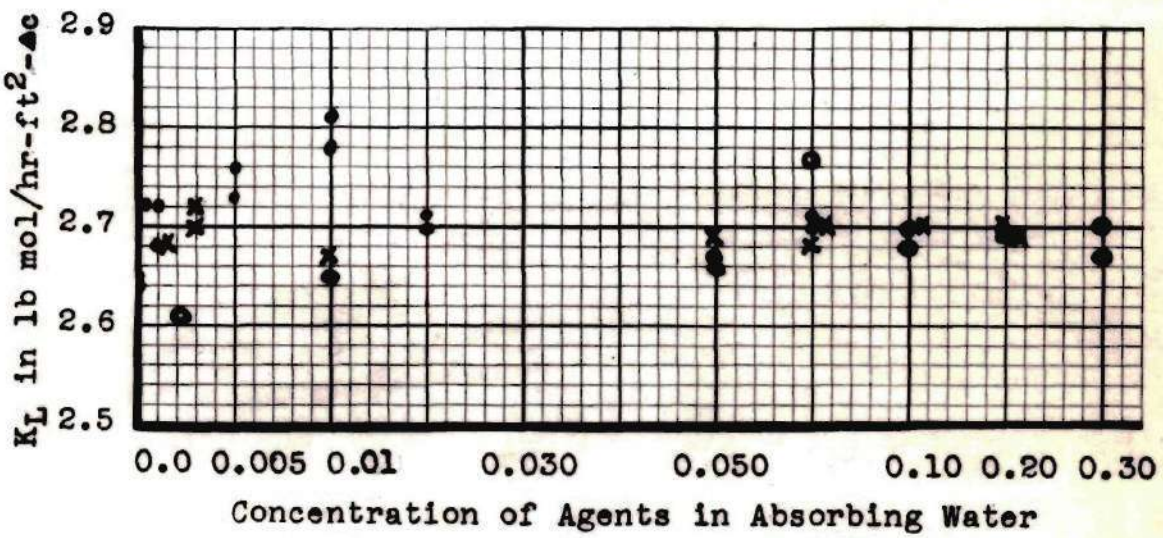


Figure 20. Absorption Coefficient for Sulfur Dioxide versus Concentration of Agents in Absorbing Water

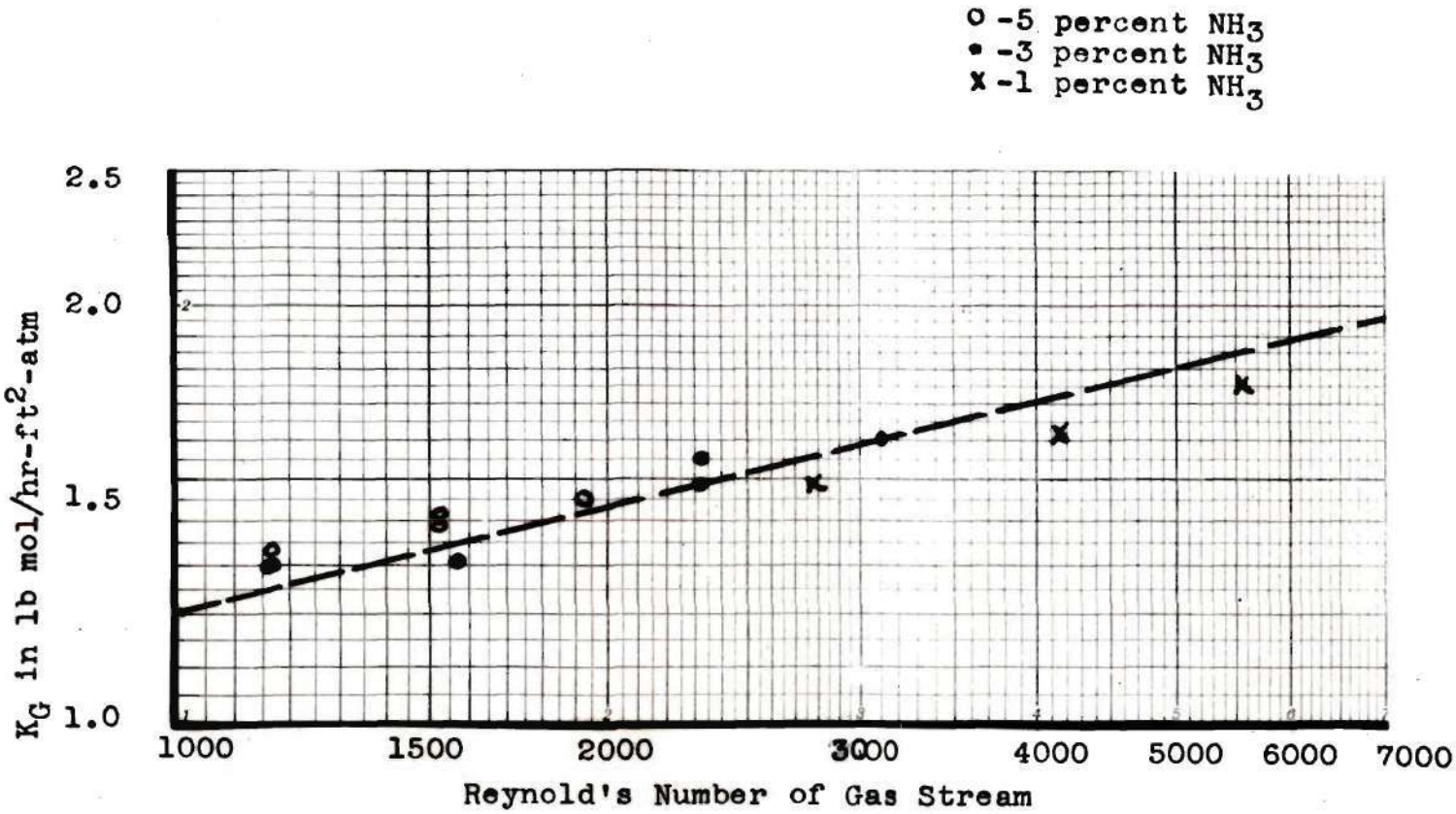


Figure 21. Absorption Coefficient for Ammonia versus Reynold's Number of Gas Stream

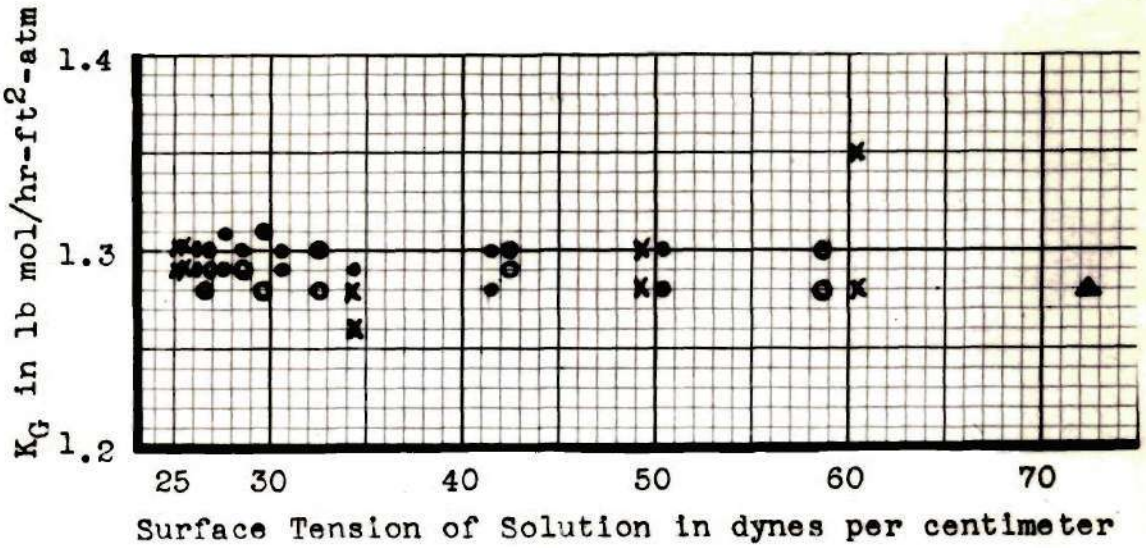


Figure 22. Absorption Coefficient for Ammonia versus Surface Tension of Absorbing Water

Both figures illustrata data for concentration of 3% NH_3 Reynold's Number of gas, 1551.8

●-Aersol OT
x-Sterox SK
●-Victawet 12
▲-Tap water

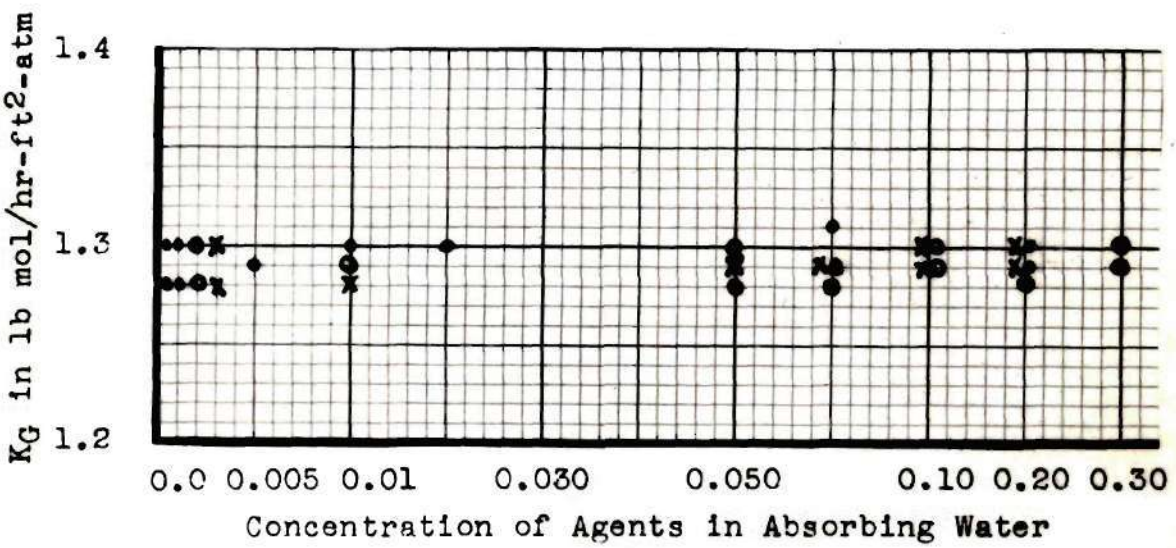


Figure 23. Absorption Coefficient for Ammonia versus Concentration of Agents in Absorbing Water

APPENDIX III

SAMPLE CALCULATION

The amount of gas absorbed.--A known amount of a standard solution of barium hydroxide was neutralized by a standard solution of hydrochloric acid and the liquid sample from the column containing carbon dioxide in water solution. Through knowledge of the amount of the acid, base, and liquid sample used and the normality of the standard acid and base solutions, the normality of the carbon dioxide sample solution was determined. The amount of carbon dioxide absorbed was then calculated.

For example:

Experimental Run Number 5

Volume of liquid sample from top of column	100 ml
Volume of barium hydroxide used with top sample.	25 ml
Volume of hydrochloric acid used to neutralize top sample	37.25 ml
Volume of liquid sample from bottom of column.	100 ml
Volume of barium hydroxide used with bottom sample . . .	50 ml
Volume of hydrochloric acid used to neutralize bottom sample.	44.40 ml
Volume of sample removed from reservoir	100 ml
Volume of barium hydroxide used with reservoir sample. .	0.0 ml
Volume of hydrochloric acid used to neutralize reservoir sample	0.0 ml
Normality of standard hydrochloric acid solution	0.04586
Normality of standard barium hydroxide solution.	0.06824

The analysis of the liquid sample from the reservoir indicated that the reservoir water was essentially neutral.

The normality of the liquid sample removed from the top of the

column was computed by a milliequivalent balance, thus,

$$(\text{ml} \times \text{N})_{HCl} + (\text{ml} \times \text{N})_{H_2CO_3} = (\text{ml} \times \text{N})_{Ba(OH)_2}$$

or,

$$N_{H_2CO_3} = \frac{(\text{ml} \times \text{N})_{Ba(OH)_2} - (\text{ml} \times \text{N})_{HCl}}{(\text{ml})_{H_2CO_3}}$$

where N is the normality of the solution

ml is the milliliters of the solution.

Substituting experimental data,

$$N_{H_2CO_3} = \frac{(25)(0.06824) - (37.25)(0.04586)}{100} = 0.00$$

Therefore, no carbon dioxide was absorbed by the liquid drops during the period of drop formation.

The normality of the bottom sample was computed by the same milliequivalent balance as previously shown. Substituting experimental values,

$$N_{H_2CO_3} = \frac{(50)(0.06824) - (44.40)(0.04586)}{100} = 1.376 \times 10^{-2}$$

Since a one normal solution of H_2CO_3 contains 1/2 of a gram mole of carbon dioxide or 22 grams per liter, the concentration of carbon dioxide in the liquid sample was,

$$(1.376 \times 10^{-2})(22/44) = 6.880 \times 10^{-3} \text{ gram moles of carbon dioxide per liter}$$

or converting, 4.295×10^{-4} pound moles of carbon dioxide per cubic foot.

Calculation of the effective area available for mass transfer.--The average number of drops per hole per minute, N_D, was determined as 100. The surface available for absorption was then calculated through use of equation (39) where,

$$\text{Total available area} = (4.47 \times 10^{-3})(N_D)^{1/3}$$

$$= 2.08 \times 10^{-2} \text{ square feet.}$$

Calculation of NA.--The liquid rate was a constant of 0.5 pounds of water per minute throughout the investigation. The rate of absorption was calculated as,

$$\frac{(4.295 \times 10^{-4})(0.5)(60)}{(62.4)(2.08 \times 10^{-2})} = 9.91 \times 10^{-3}$$ pound moles of carbon dioxide per hour-square foot.

Calculation of the absorption coefficient.--From simple absorption theory,

$$N_A = K_L (\Delta N)_{lm}$$

where N_A is the amount of gas absorbed per unit time per unit area,

K_L is the over-all absorption coefficient,

$(\Delta N)_{lm}$ is the logarithmic mean of the driving forces $(\Delta N)_B$ at the bottom of the column and $(\Delta N)_T$ at the top of the column and (ΔN) equals to $(N_e - N)$. Here N is expressed in mole fractions.

The concentration of carbon dioxide in the bottom liquid sample was found to be 6.88×10^{-3} gram moles per liter which was expressed in mole fraction form as,

$$N_B = \frac{6.88 \times 10^{-3}}{55.55} = 1.238 \times 10^{-4}, \text{ mole fraction.}$$

The equilibrium mole fraction at the bottom of the column was calculated as follows:

The gas entering the column contained 80 per cent carbon dioxide by volume and was at a temperature of 79 degrees Fahrenheit and a pressure of 742 mm of mercuty. Henry's law constant under these conditions is 1.28×10^6 mm of mercury per mole of carbon dioxide per mole of solution. The equilibrium mole fraction was then,

$$(N_e)_B = \frac{(0.80)(742)}{1.28 \times 10^6} = 4.638 \times 10^{-4}, \text{ mole fraction.}$$

Therefore,

$$(\Delta N)_B = (N_e)_B - (N)_B = (4.638 - 1.238) \times 10^{-4}$$

$$= 3.400 \times 10^{-4}$$

At the top of the column the procedure was as follows:

The gas rate was 524.200×10^{-4} pound moles per hour entering the column of which 80 per cent or $(0.80)(524.200 \times 10^{-4})$, 419.300×10^{-4} pound moles per hour was carbon dioxide. The amount of carbon dioxide absorbed in pound moles per hour was obtained from previous calculations by multiplying N_A, the pound moles of carbon dioxide absorbed per hour-square foot times the effective area available for absorption in square feet as follows,

Pound moles carbon dioxide absorbed per hour = $(N_A)(A)$

or, $(9.91 \times 10^{-3})(2.08 \times 10^{-2}) = 2.065 \times 10^{-4}$ pound moles.

Therefore, carbon dioxide was leaving the column at a rate of $(419.300 - 2.065) \times 10^{-4}$ or 417.235×10^{-4} pound moles per hour.

Air was leaving the top of the column at a rate of $(1.00 - 0.80)(419.300 \times 10^{-4})$ or 104.900 pound moles per hour. Therefore, the mole fraction of carbon dioxide in the gas leaving the top of the column was,

$$\frac{417.235 \times 10^{-4}}{(417.235 - 104.900) \times 10^{-4}} = 0.7991$$

from which the equilibrium mole fraction was calculated as,

$$(N_e)_T = \frac{(0.7991)(742)}{1.28 \times 10^6} = 4.632 \times 10^{-4}$$

The mole fraction of carbon dioxide in the liquid at the top of the column was negligible; $(\Delta N)_T$ was thus,

$$(\Delta N)_T = (N_e)_T - (N)_T = (N_e)_T = 4.632 \times 10^{-4}$$

The logarithmic mean driving force was then computed as,

$$(\Delta N)_{lm} = \frac{(\Delta N)_T - (\Delta N)_B}{\ln \frac{(\Delta N)_T}{(\Delta N)_B}} = \frac{4.632 \times 10^{-4} - 3.400 \times 10^{-4}}{\ln \left(\frac{4.632 \times 10^{-4}}{3.400 \times 10^{-4}} \right)}$$

$$= \frac{1.232 \times 10^{-4}}{\ln(1.362)} = \frac{1.232 \times 10^{-4}}{0.3085}$$

$$= 3.999 \times 10^{-4}$$

The over-all coefficient was, therefore,

$$K_L = \frac{N_A}{(\Delta N)_{lm}} = \frac{0.991 \times 10^{-2}}{3.999 \times 10^{-4}}$$

$$= 24.78 \text{ pound moles per hour-square foot- } \Delta N$$

To express the driving force in terms of concentration expressed as pound moles per cubic foot, since one pound mole of solution occupies 0.2883 cubic feet, the over-all absorption coefficient was then,

$$K_L = 24.78 \times 0.2883 = 7.144 \text{ pound moles per hour-square foot- } \Delta c.$$

Correction of the absorption coefficient for temperature.--For runs not at 80° Fahrenheit, temperature corrections were introduced following the method of Sherwood and Holloway (8) where,

$$K_L = me^{nT}$$

m and n are constants and T is the temperature in degrees Centigrade. According to Sherwood and Holloway, n = 0.021, thus,

$$\frac{(K_L)_{T_1}}{(K_L)_{T_2}} = e^{n(T_1 - T_2)}$$

or K_L corrected to 80 degrees Fahrenheit is (7.114)(1.012)

$$K_L \ (80^\circ F.) = 7.23$$

APPENDIX IV

GIBBS FILM CALCULATION

When a Gibbs surface is postulated, it must be known for the drop size used whether or not there are a sufficient number of molecules of the surface active agent in the system to provide the molecules for the Gibbs surface without seriously affecting the bulk concentration.

When spherical drops of the size used in this investigation are formed, it is easily shown that the bulk concentration is not appreciably affected by the creation of the new surface of the drop.

The diameter of the drops formed on the distributor tap in this investigation by a 0.1 weight per cent solution of Aersol OT was found to be 0.162 inches. The surface area of a drop may be computed from the relation,

$$\frac{\pi(0.162)^2}{144} = 5.71 \times 10^{-4} \text{ square feet}$$

The excess surface concentration at this bulk concentration, 0.10 weight per cent, from Table I-A of the Appendix, is 3.12×10^{-10} pound moles per square foot. The pound moles of Aersol in the surface film in excess of the normal contribution of the bulk concentration is then computed as,

$$(5.71 \times 10^{-4} \text{ ft}^2)(3.12 \times 10^{-10} \text{ lb.moles/ft}^2)$$

$$\text{or } 1.78 \times 10^{-13} \text{ pound moles}$$

The actual surface concentration is the sum of the Gibbs excess surface concentration and the contribution of the liquid bulk concentration. Postulating an equal distribution of the molecules in the liquid bulk, the volume fraction of Aersol may be expressed as,

$$\frac{\text{grams Aersol}}{\text{cc solution}} \cdot \frac{1}{\text{density of Aersol (gms/cc)}} = \text{Volume fraction}$$

Then since volume fraction equals surface fraction, dividing the surface fraction of Aersol by the area per molecule, assumed as 30 square Angstrom units for this illustration, and the number of molecules per mole and then converting to units of pound moles per square foot, the following is obtained:

$$\text{pound moles/square foot} = 0.661 \times 10^{-15} \text{ (wt conc Aersol)}$$

$$= 0.661 \times 10^{-15} \ (0.001)$$

$$= 6.61 \times 10^{-19}$$

which is negligible in comparison to the Gibbs surface contribution.

The total moles of Aersol in the drop is,

$$\left(\frac{\pi}{6} \frac{(0.162)^3}{1728} \text{ ft}^3\right) \left(62.4 \frac{\text{pounds}}{\text{ft}^3}\right) \left(0.001 \frac{\text{lbs Aersol}}{\text{lb } H_2O}\right)$$

$$\left(\frac{1}{\text{mol wt Aersol}}\right)$$

or, total pound moles = 1.80×10^{-10}

Consequently, the removal of the surface molecules from the liquid bulk

will cause no significant change in the bulk concentration.

A calculation of the theoretical amount of the surface covered assuming that the Aersol molecules are in a perpendicular position on the liquid surface giving an effective cross sectional area of 30 square Angstrom units per molecule, the Gibbs excess surface concentration molecules would cover 27.4 per cent of the surface, while the bulk contribution would cover only 0.05 per cent of the surface, a negligible amount.

At a bulk concentration, 0.05 weight per cent Aersol, which yields a maximum for the Gibbs excess surface concentration, use of the previous methods of calculations gives the following results:

Aersol OT in the Gibbs surface	4.14×10^{-13} lb moles
Aersol OT in the liquid bulk	1.13×10^{-10} lb moles
Percent surface covered by the Gibbs surface molecules	64
Percent surface covered by the bulk contribution	Negligible

In consideration of a wetted wall column study using water with added Aersol at a bulk concentration of 0.10 weight per cent, the pound moles of Aersol OT needed for the formation of one square inch of a Gibbs surface would be,

$$(6.94 \times 10^{-3} ft^2)(3.12 \times 10^{-10} \text{ lb moles}/ft^2)$$

$$= 2.16 \times 10^{-12} \text{ lb moles}$$

However, the pound moles available will be a function of the depth of the surface, assuming a maximum wall liquid thickness of 0.1 inches, the pound moles of Aersol available from the bulk is,

$(5.79 \times 10^{-5}\ ft^3)(62.4\ lbs/ft^3)(0.001\ lbs\ Aersol/lb\ H_2O)$

$(1/mol\ wt)$

or $= 8.15 \times 10^{-9}$ pound moles of Aersol

Thus, even if the wall liquid thickness were only 0.01 inches, the available Aersol in the bulk would be 8.15×10^{-10} pound moles and the removal of the amount required for the Gibbs surface is still negligible.

At 0.05 weight per cent Aersol, the pound moles in the Gibbs surface is 4.33×10^{-12}. The amount available in the wall liquid is 4.08×10^{-9} pound moles for a wall liquid thickness of 0.1 inches or 4.08×10^{-10} pound moles for a wall liquid thickness of 0.01 inches, thus, sufficient Aersol is still available.

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